

Development of coated conductors based on IBAD MgO

Paul Arendt - MgO template development for coated conductors using ion-beam assisted deposition

Steve Foltyn - High current YBCO on IBAD MgO by pulsed laser deposition

\$1850 K, 6.0 FTE

*Superconductivity Technology Center
Los Alamos National Laboratory*

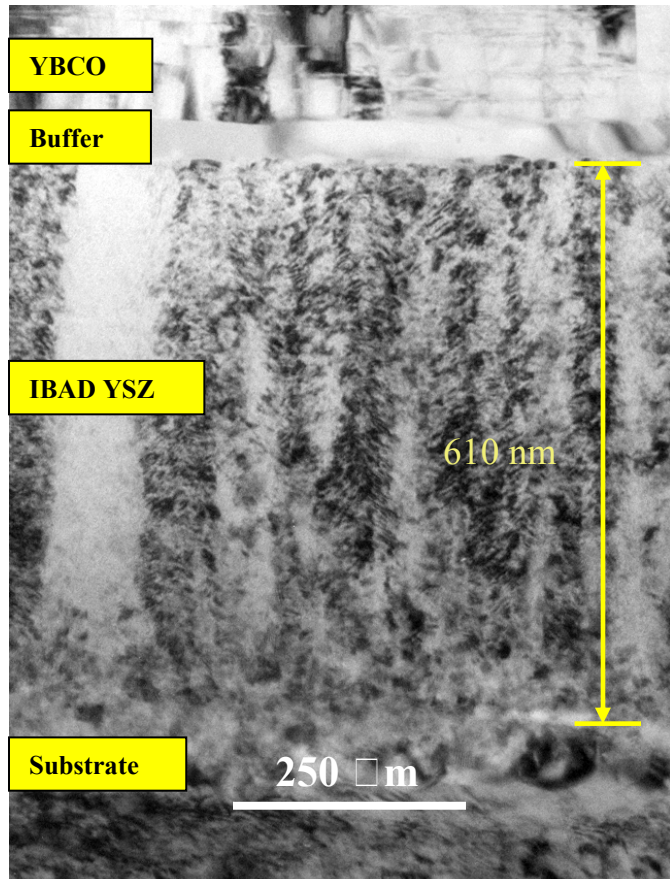
MgO template development for coated conductors using ion-beam assisted deposition

Paul Arendt, Steve Foltyn, Ray DePaula, Randy Groves,
Terry Holesinger, Quanxi Jia, Sascha Kreiskott
Vladimir Matias, Liliana Stan, Igor Usov, Haiyan Wang

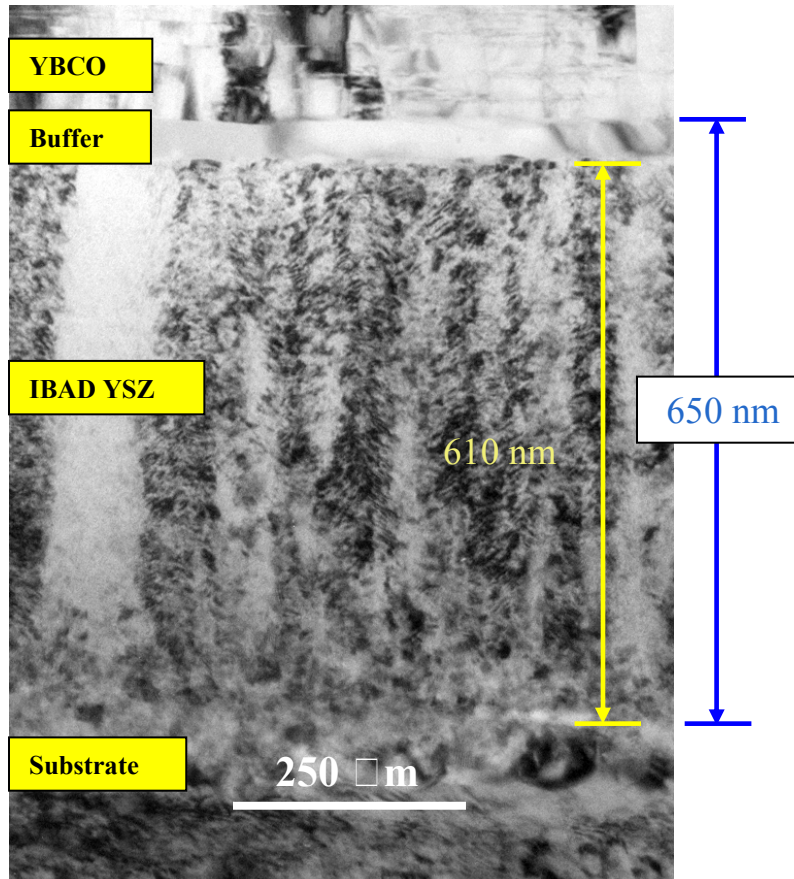
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1. Substrate cation diffusion barrier
2. IBAD MgO texture optimization
 - a) nucleation layer
 - b) assist ion beam divergence

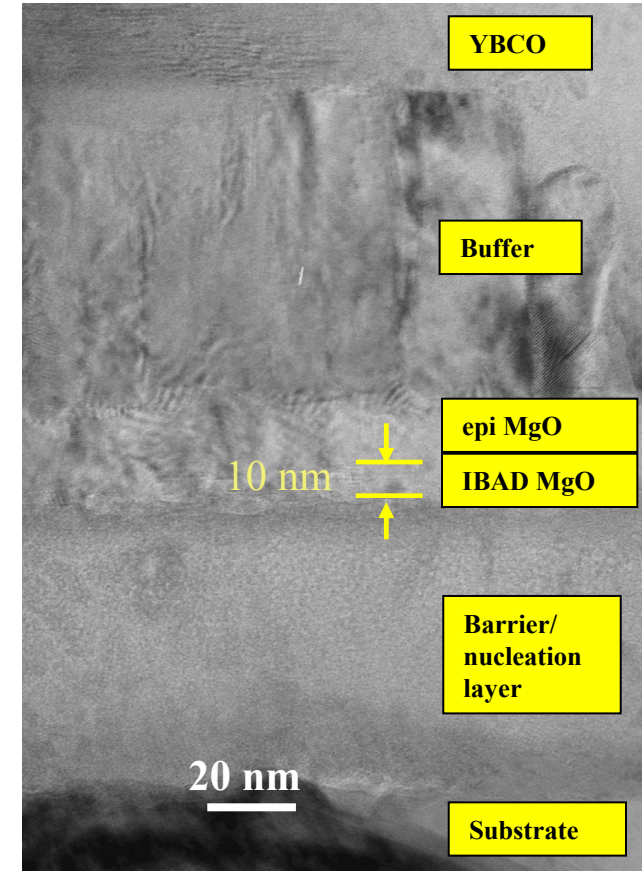
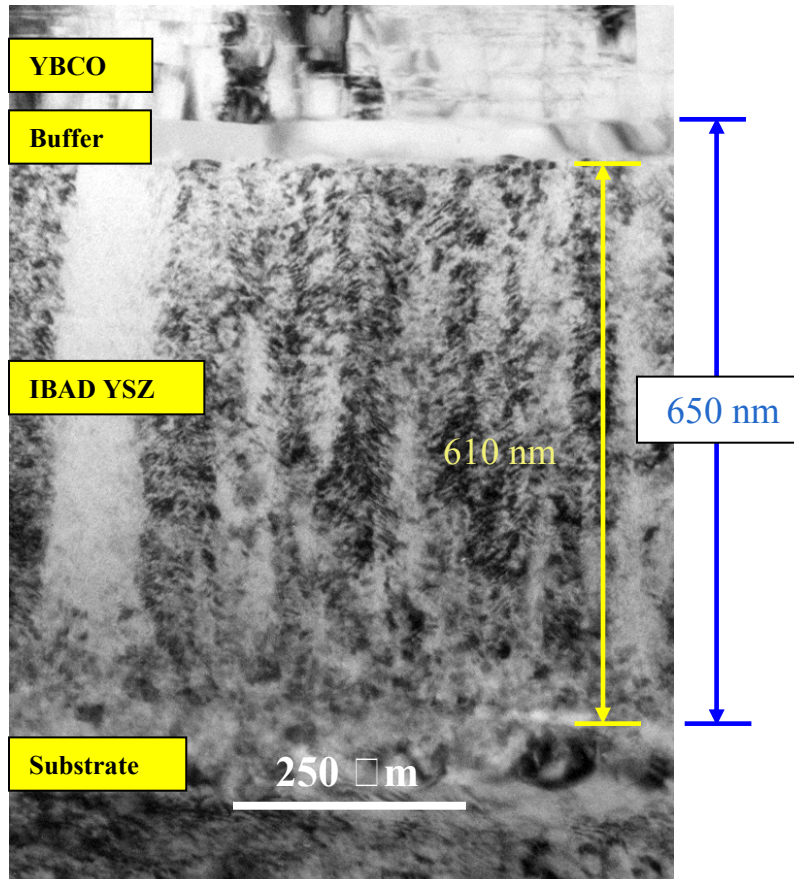
Thickness requirement of IBAD YSZ prompted our program to investigate IBAD MgO



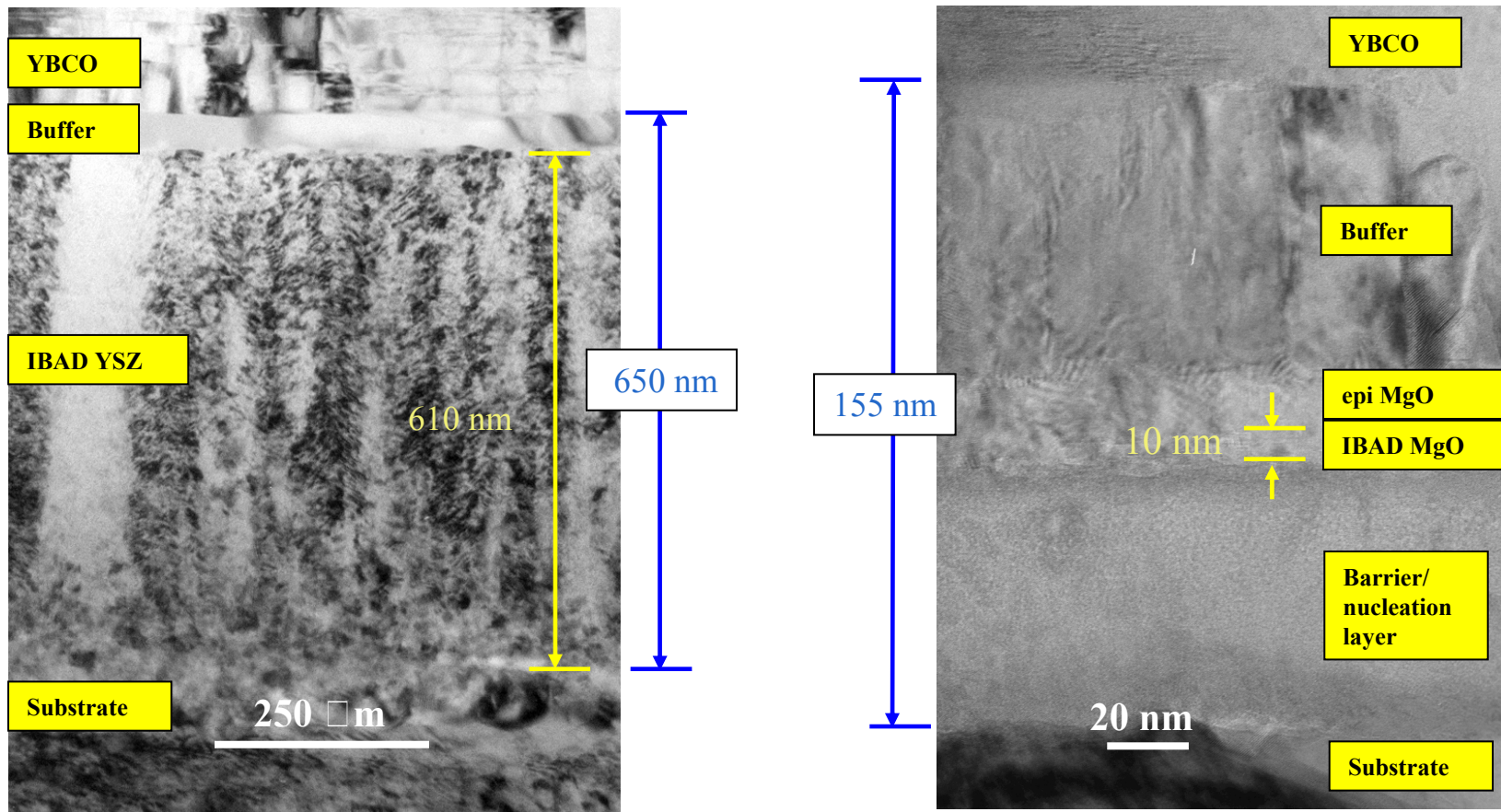
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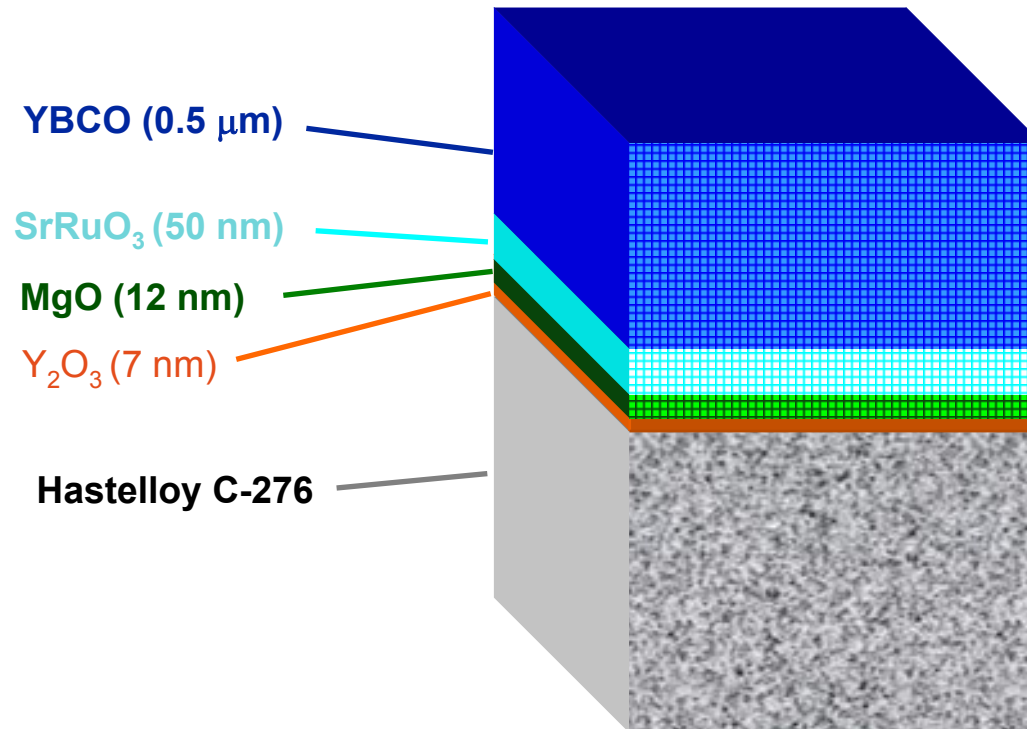
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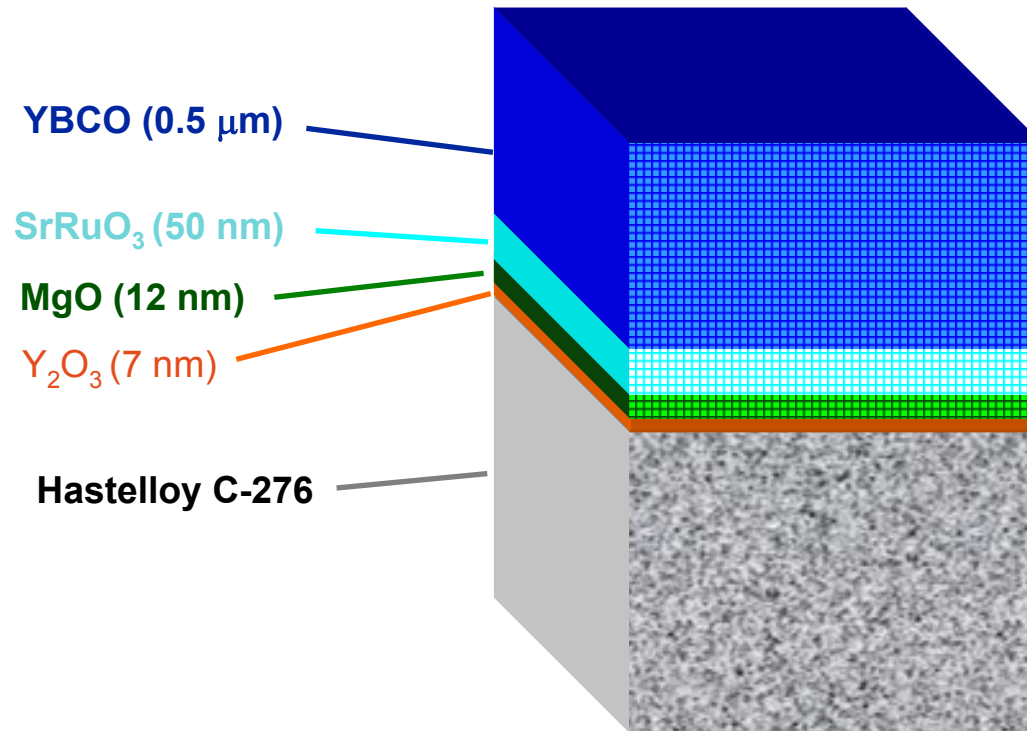
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Why have a barrier layer?

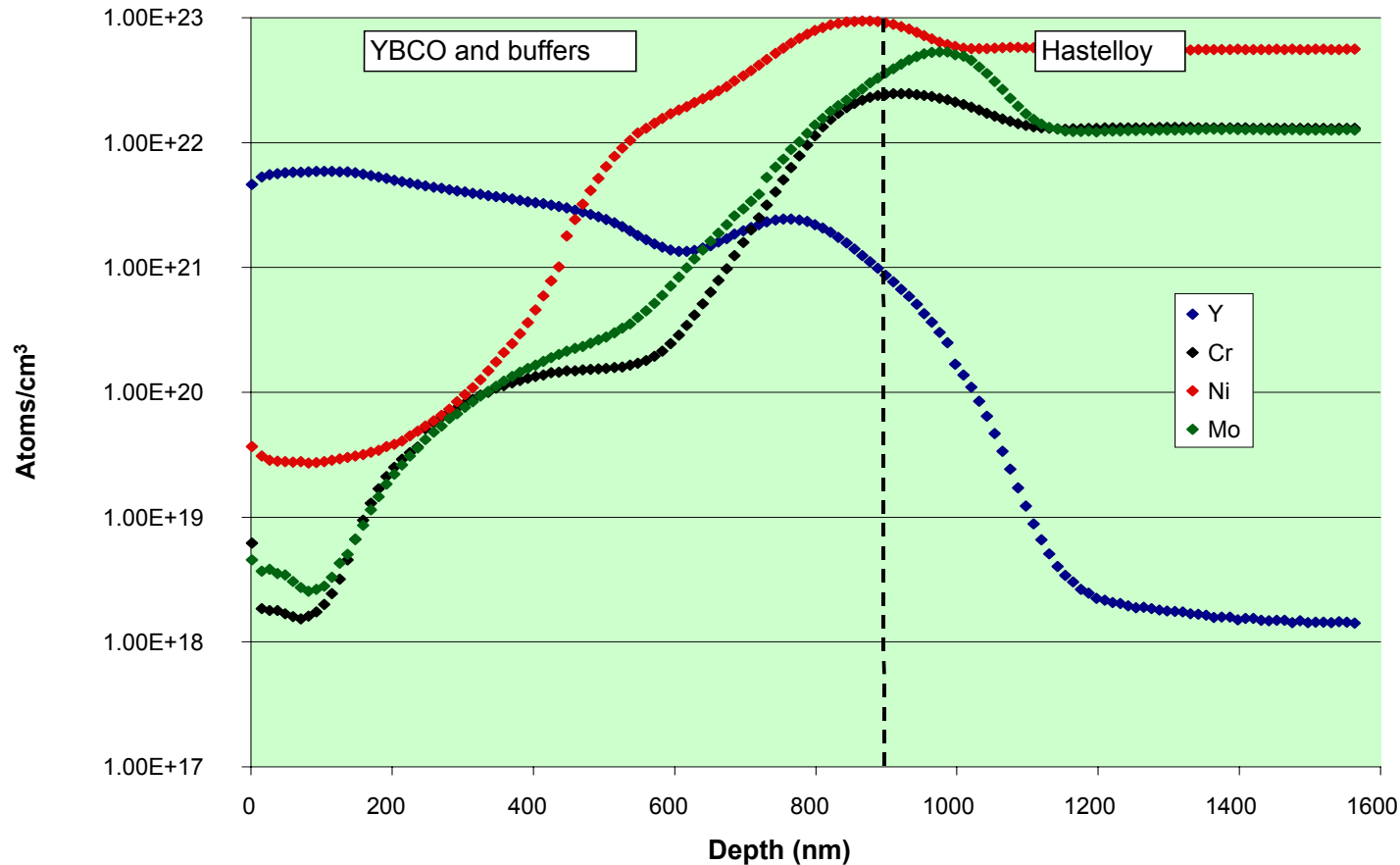


Why have a barrier layer?



$$T_c < 75K$$

SIMS depth profile of selected cations



YBCO/SRO/MgO/Y₂O₃/Hastelloy

What alternatives may prevent cations from poisoning the YBCO?

Substrate cation diffusion is thought to be resolved for IBAD YSZ with its thicker template/buffer architecture

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e.g. at normal YBCO processing temperatures, diffusion coefficients in Al_2O_3 for O and various substrate cations (Fe, Cr) tend to be much lower than for many other common oxides.

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YBCO (0.5 μm)

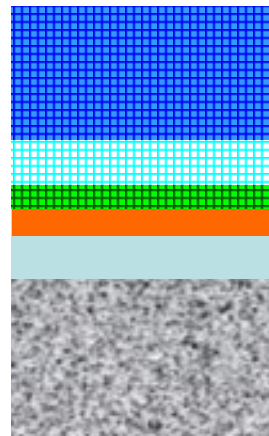
SrRuO₃ (50 nm)

MgO (12 nm)

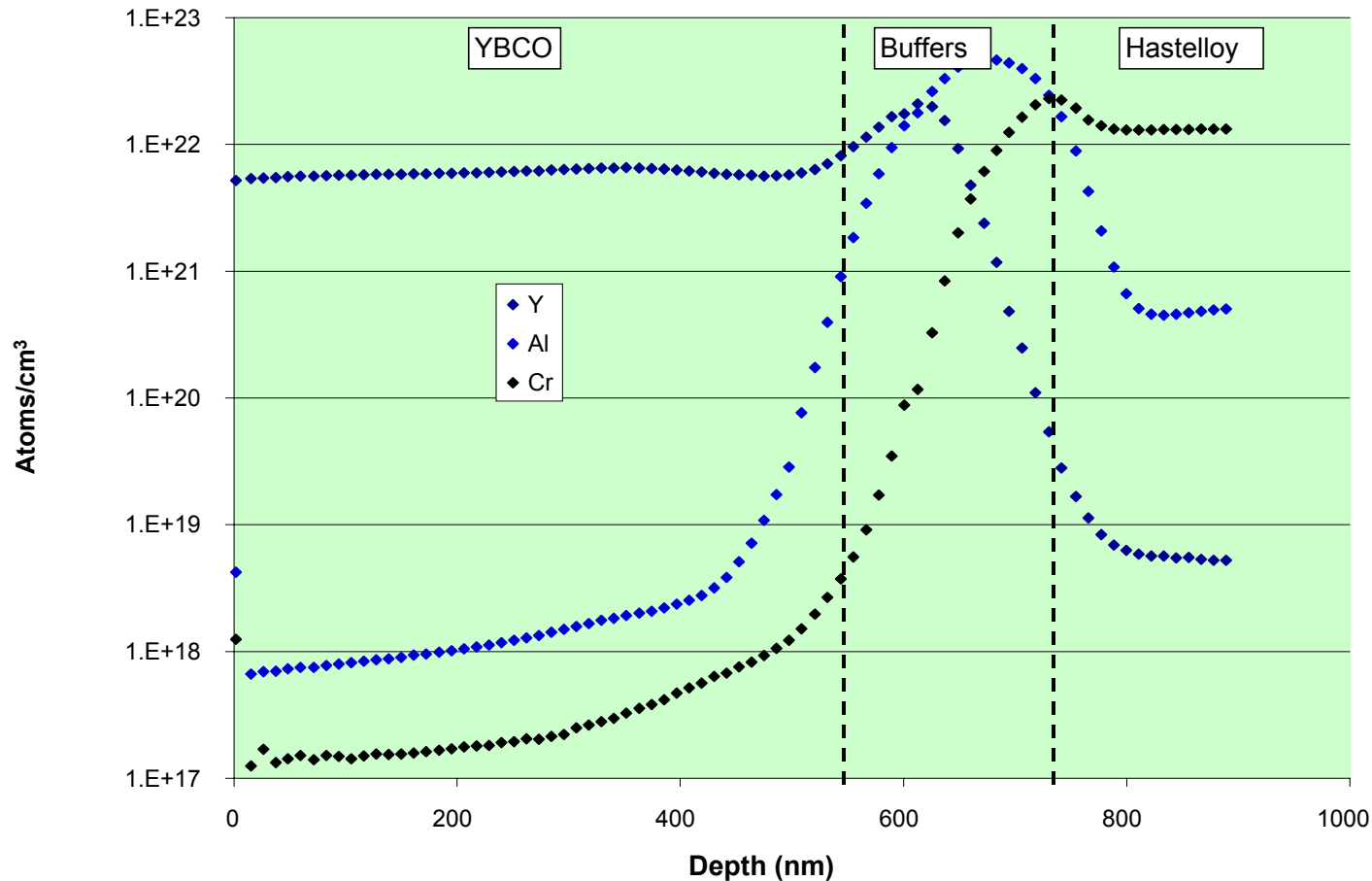
Y₂O₃ (7 nm)

Al₂O₃ (80 nm)

Hastelloy C-276

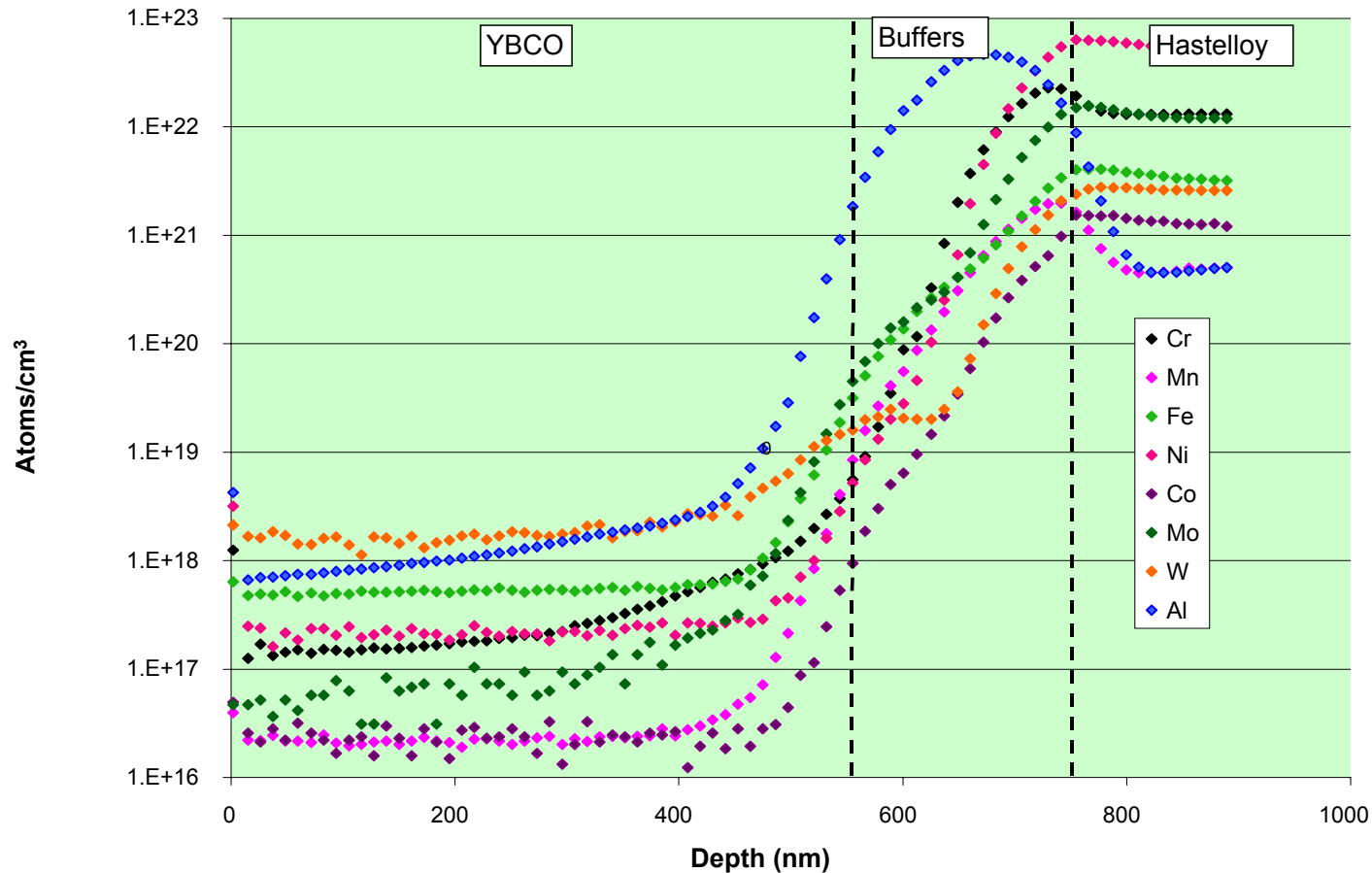


SIMS depth profile of selected cations with alumina layer deposited on substrate



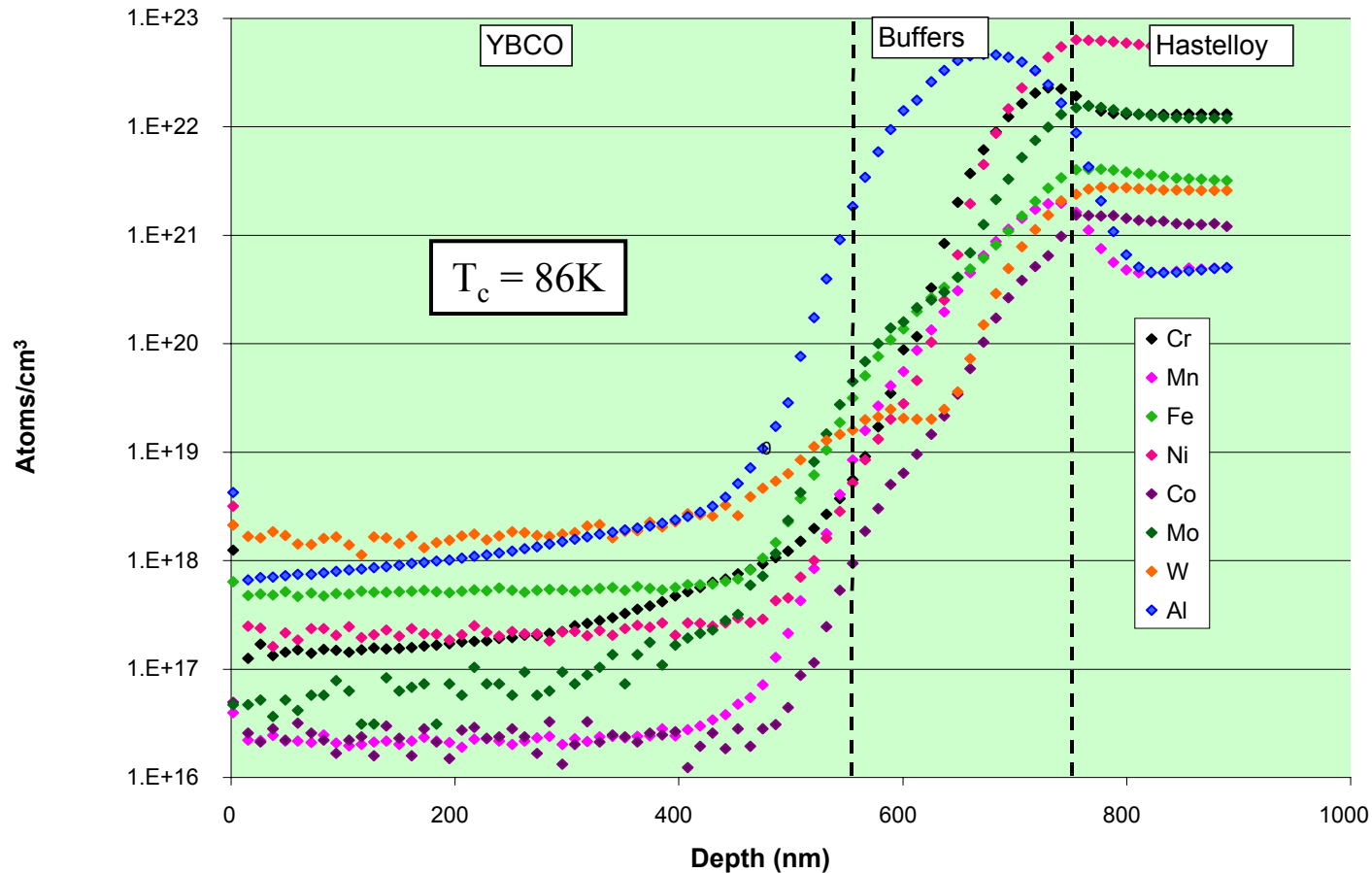
YBCO/SRO/MgO/Y₂O₃/Al₂O₃/Hastelloy

All the substrate cations are impeded from diffusing into the YBCO



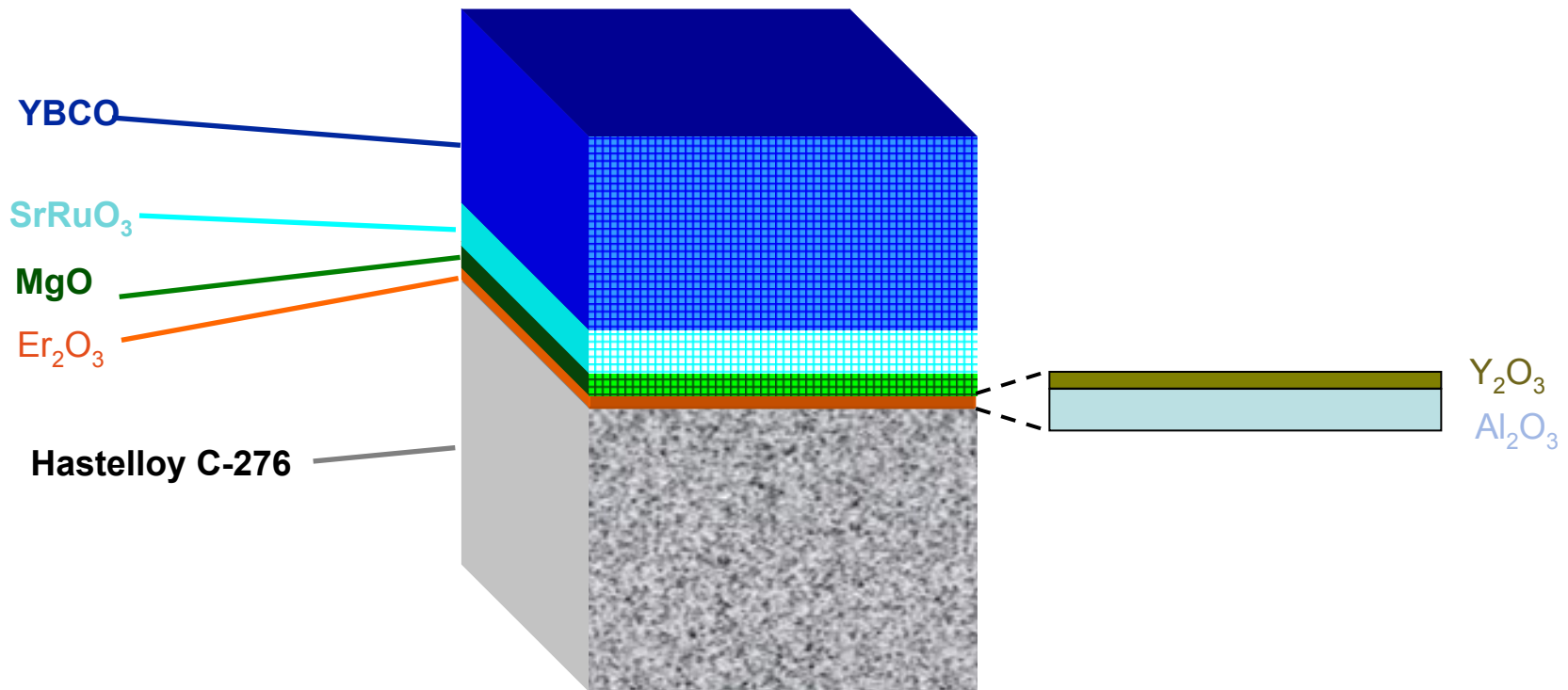
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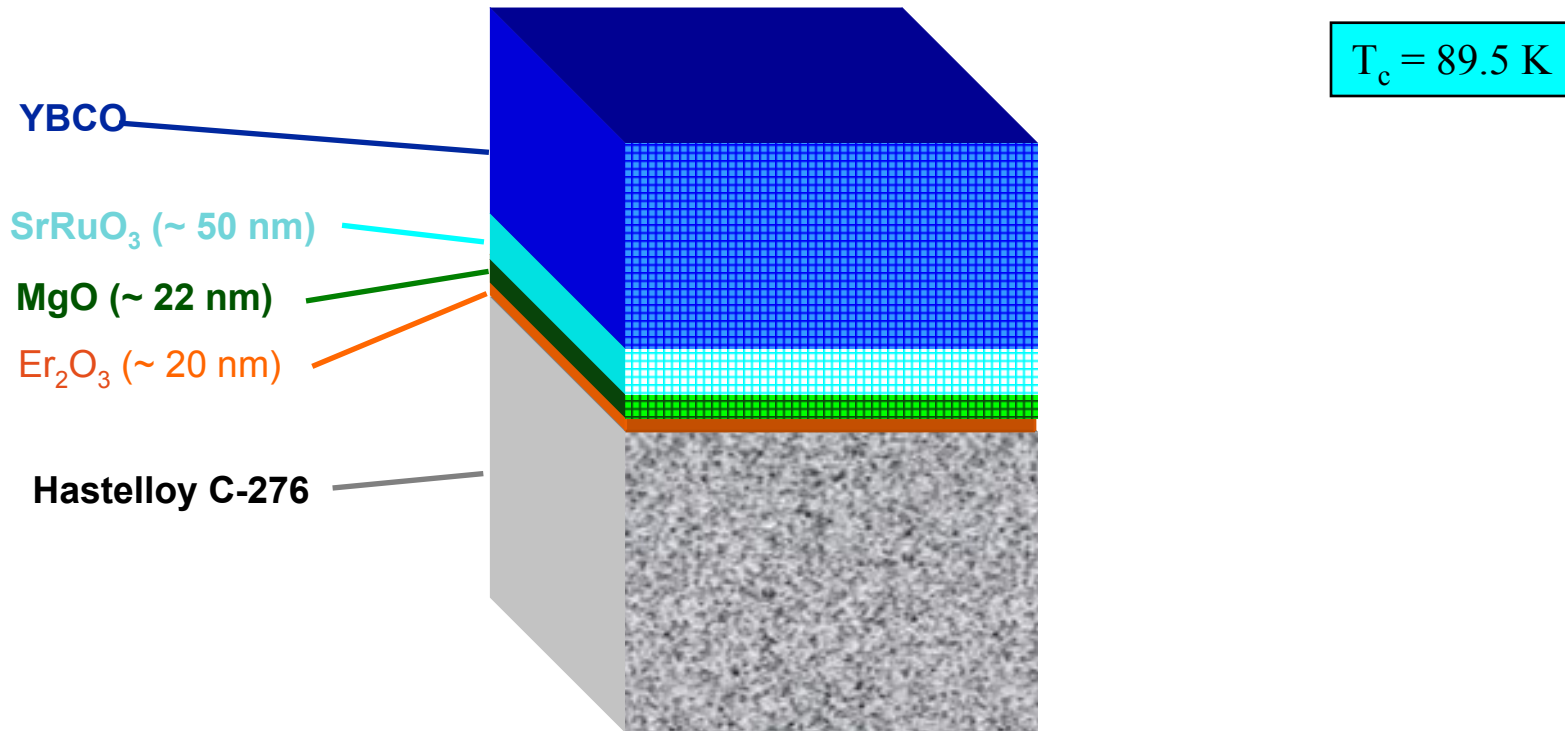


YBCO/SRO/MgO/Y₂O₃/Al₂O₃/Hastelloy

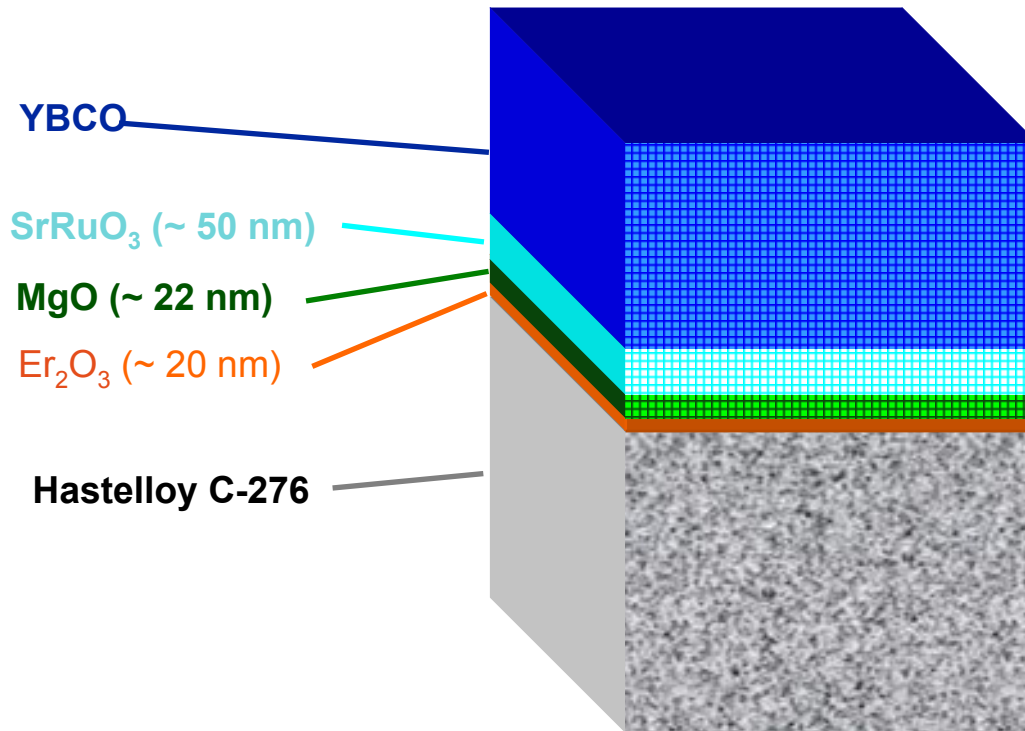
Another approach for MgO templates is to combine the barrier and nucleation layers



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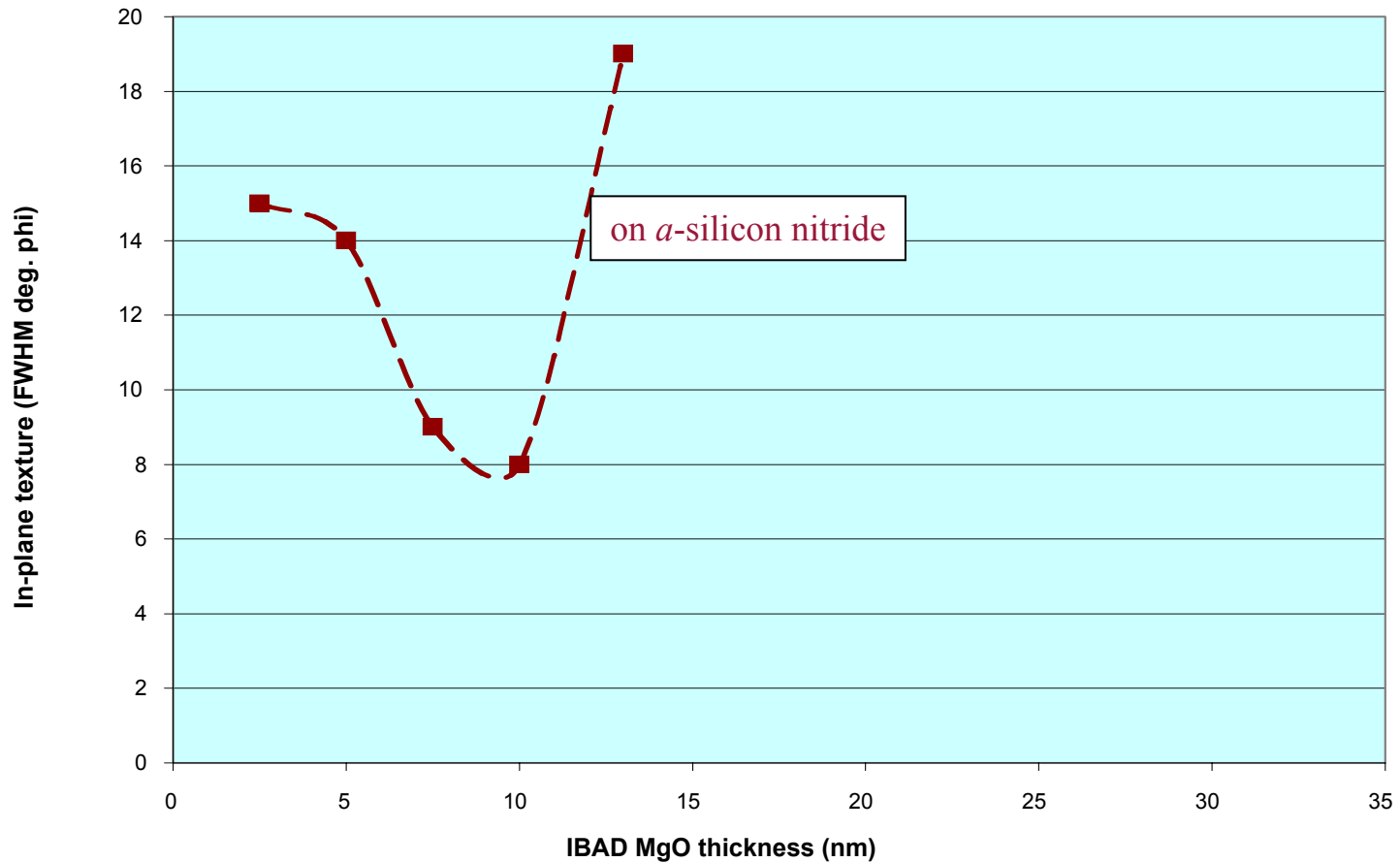
$$T_c = 89.5 \text{ K}$$

$$J_c(75\text{K}, \text{sf}) = 2.3 \text{ MA/cm}^2$$

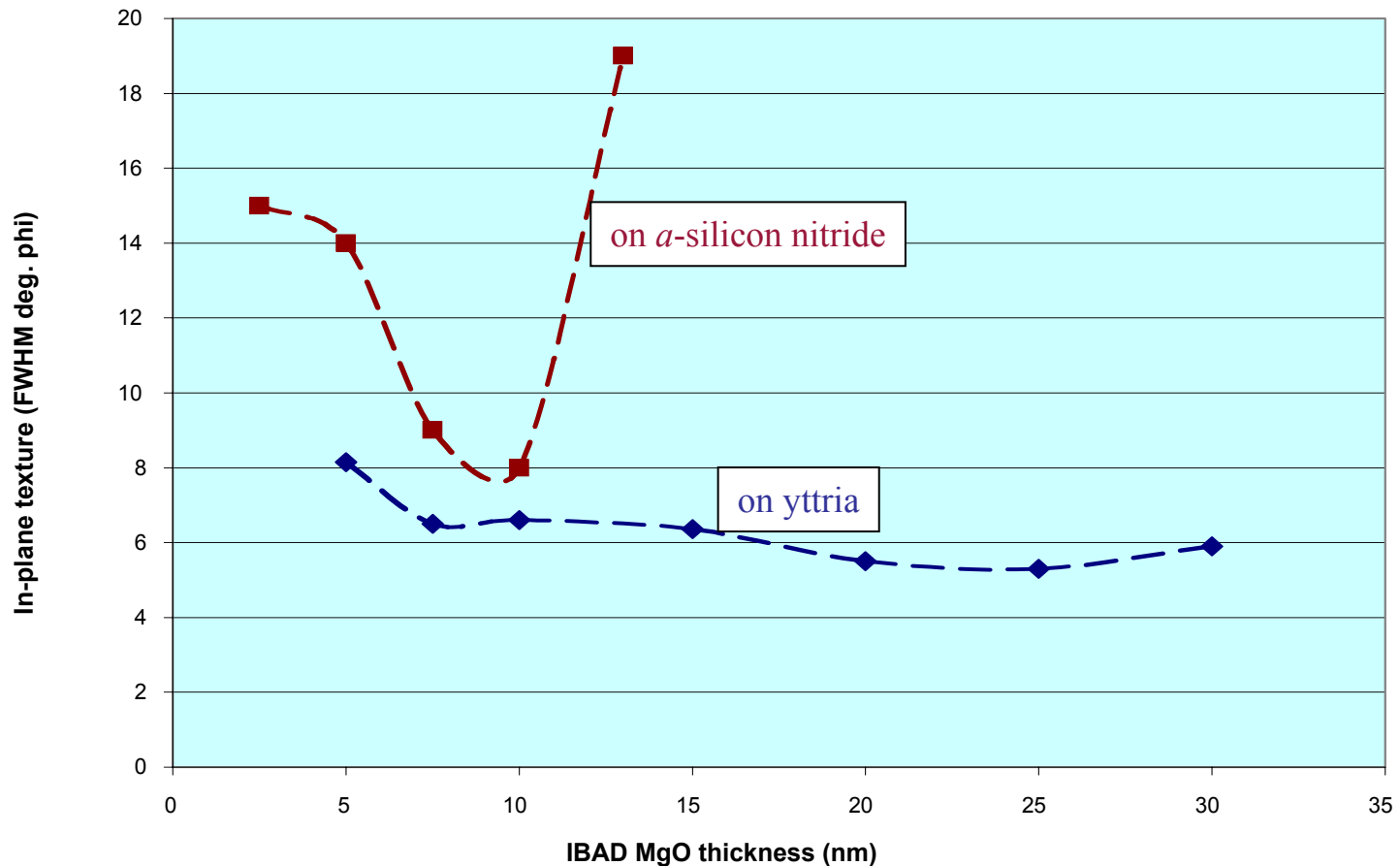
(thickness = 1.7 μm)

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1. Substrate cation diffusion barrier
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 - a) nucleation layer
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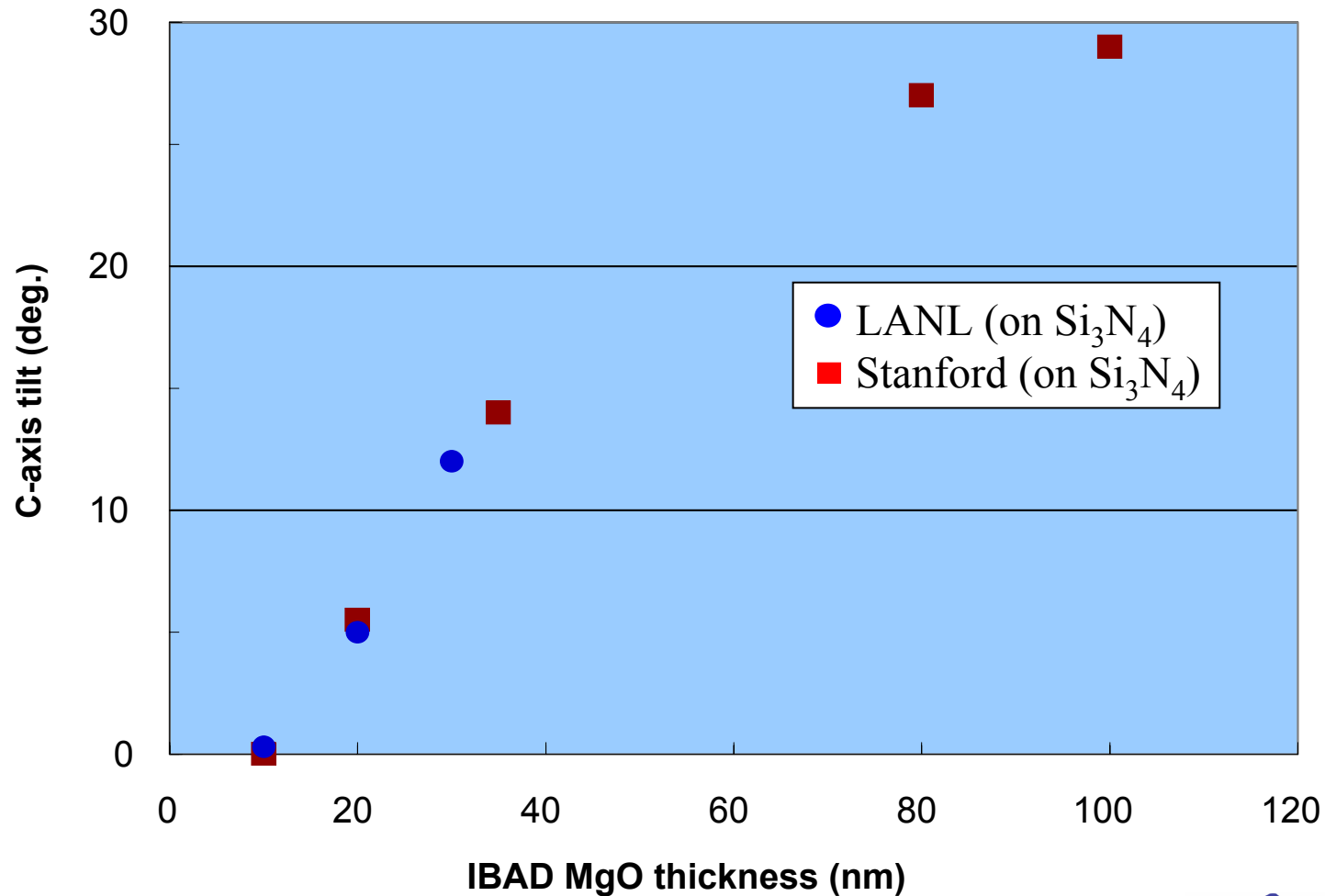
IBAD MgO texture vs. thickness processing window is extended when deposited on yttria



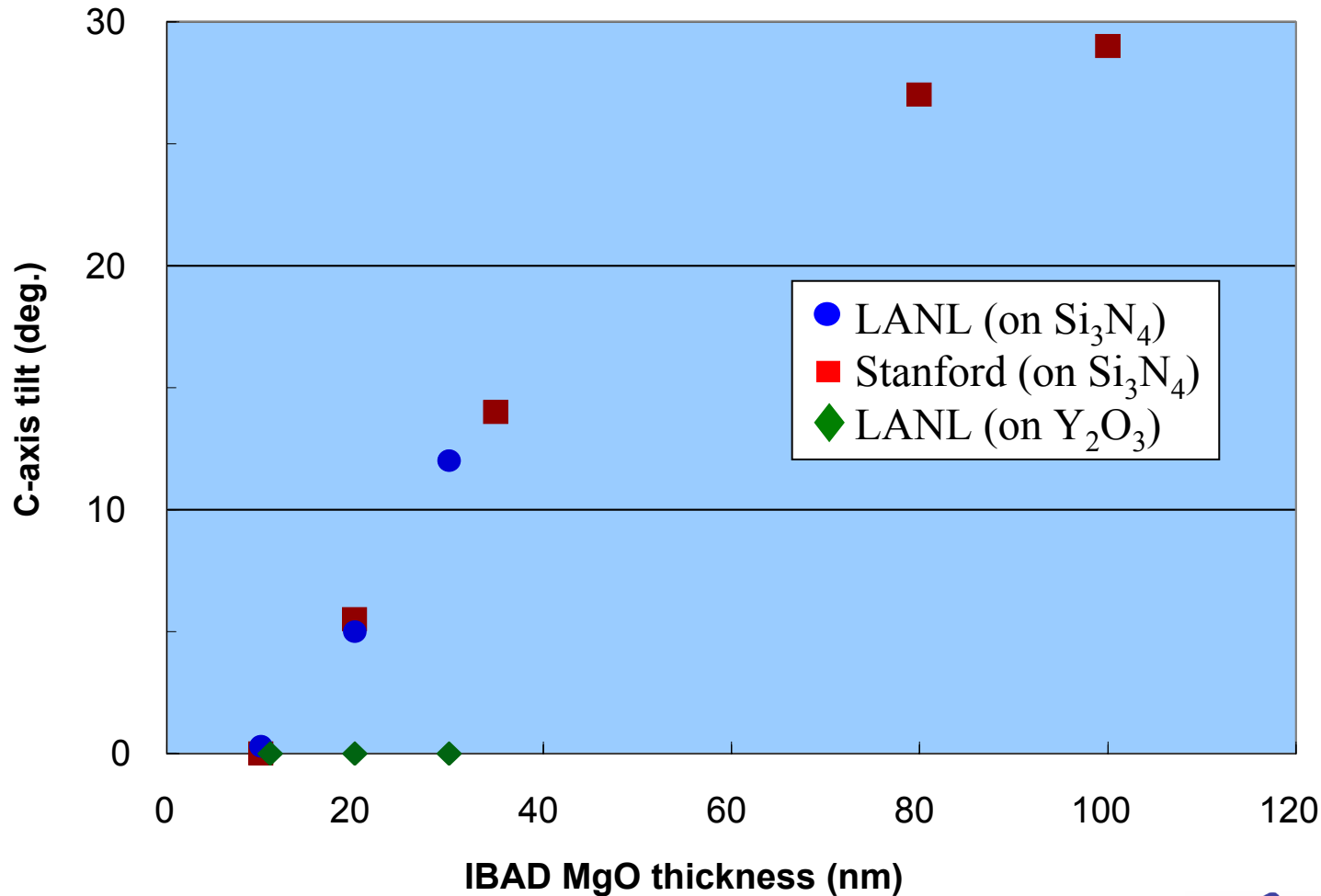
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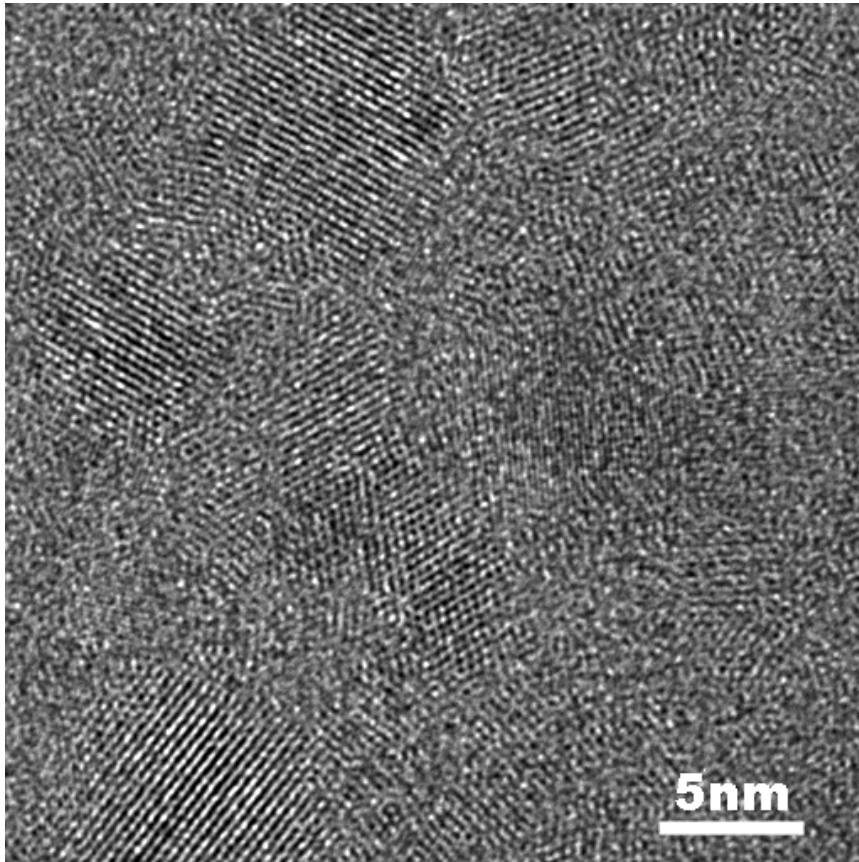
C-axis tilt for IBAD MgO on Si_3N_4 believed to be reason for texture degradation when thickness $> 100 \text{ \AA}$



No c-axis tilt observed for IBAD MgO on Y_2O_3 (up to 300 Å)

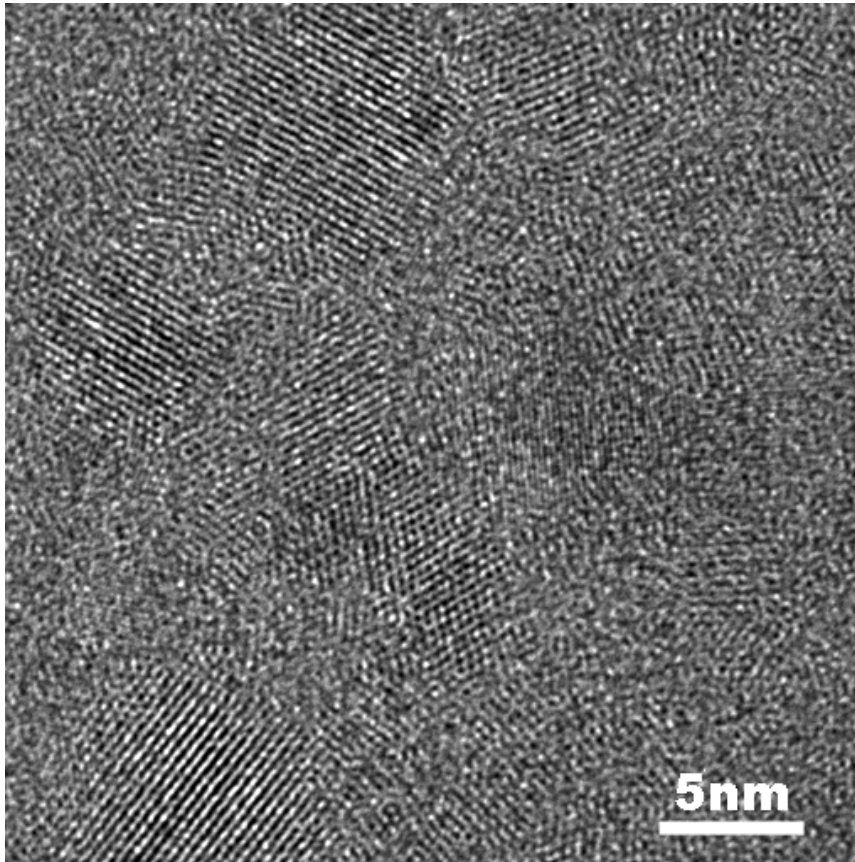


Plan view HRTEM reveals the yttria nucleation layer to be nanocrystalline

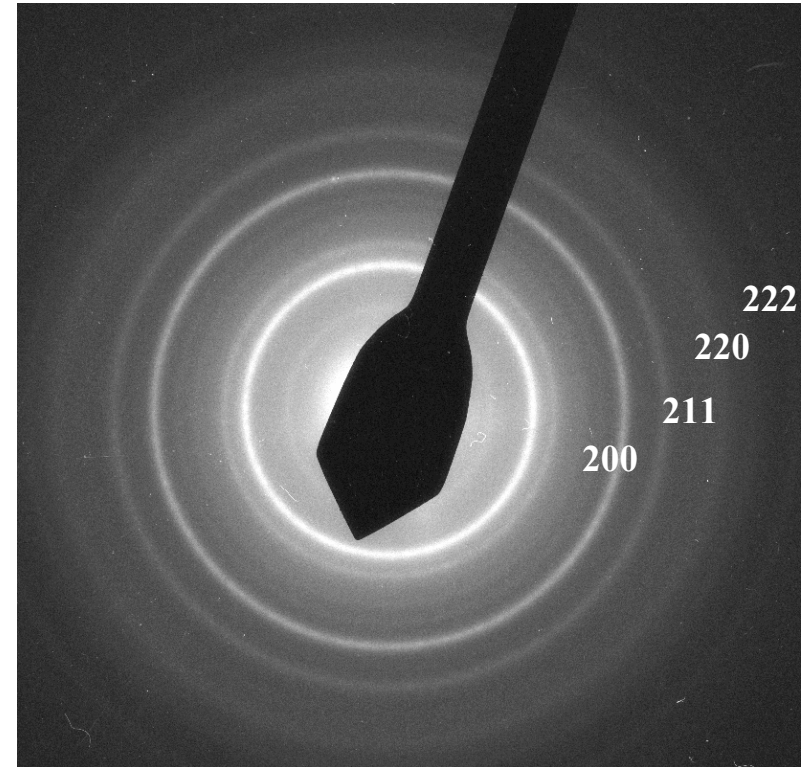


10 nm thick Y₂O₃ film

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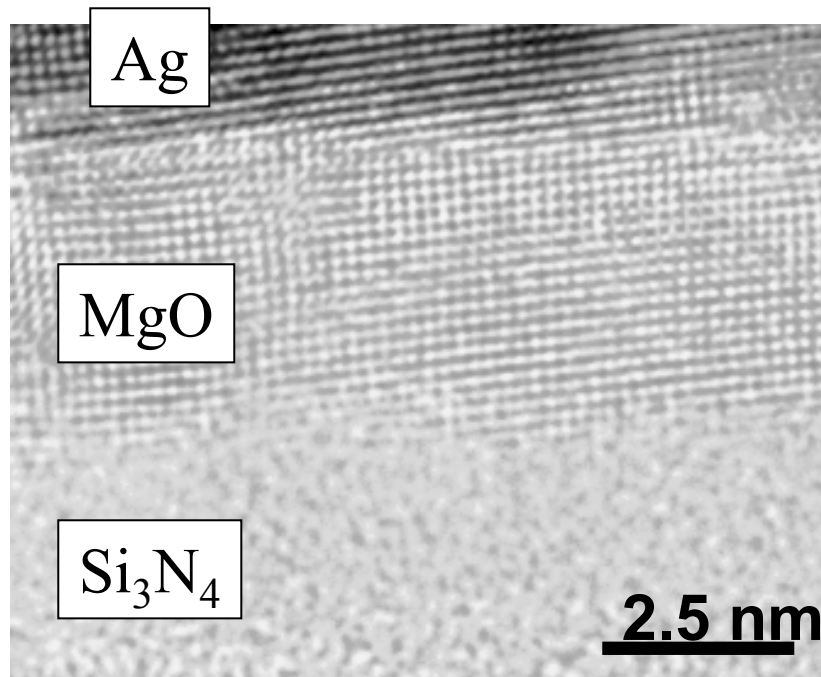


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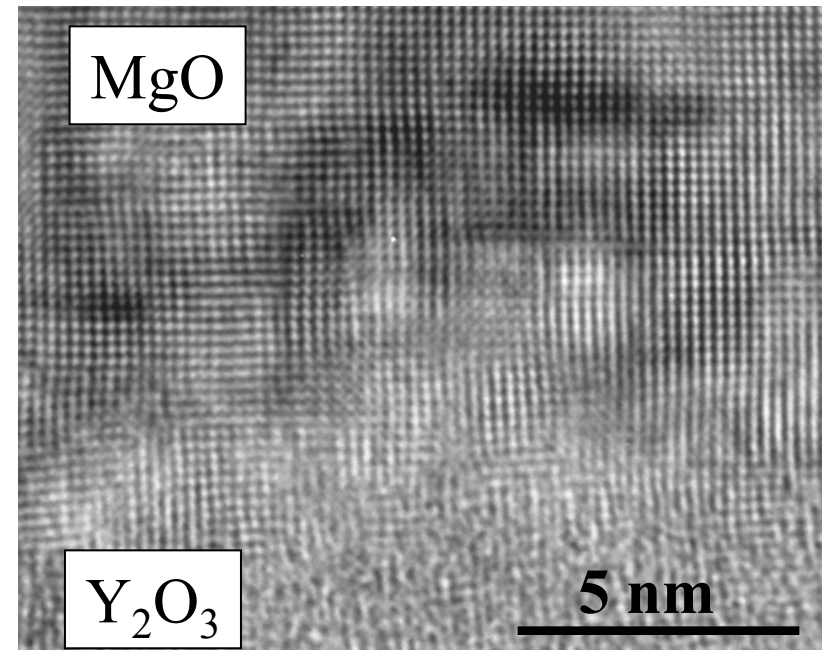
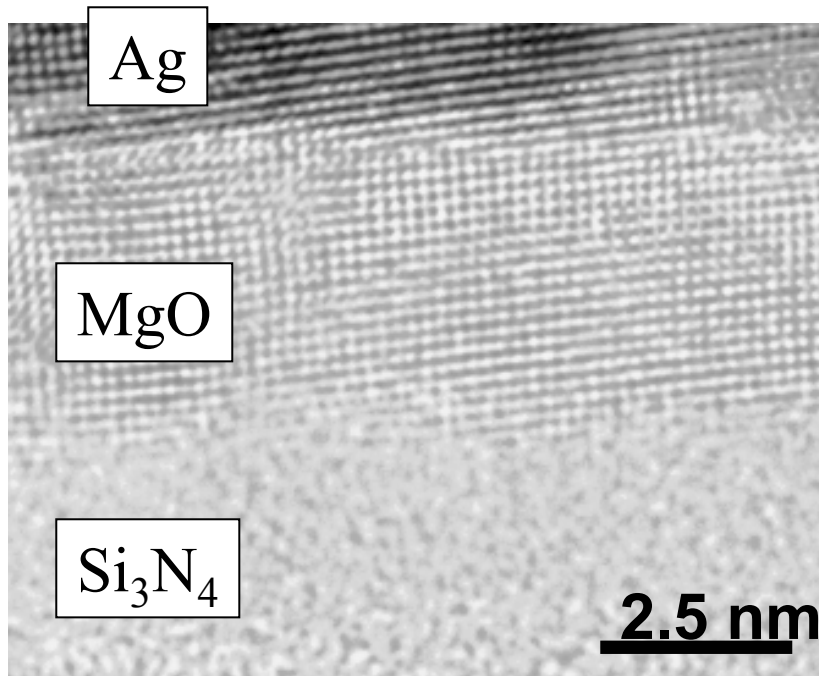


SAD pattern shows film is randomly oriented

Comparison of HRTEM cross sections reveal differing growth characteristics for IBAD MgO on the two nucleation layers

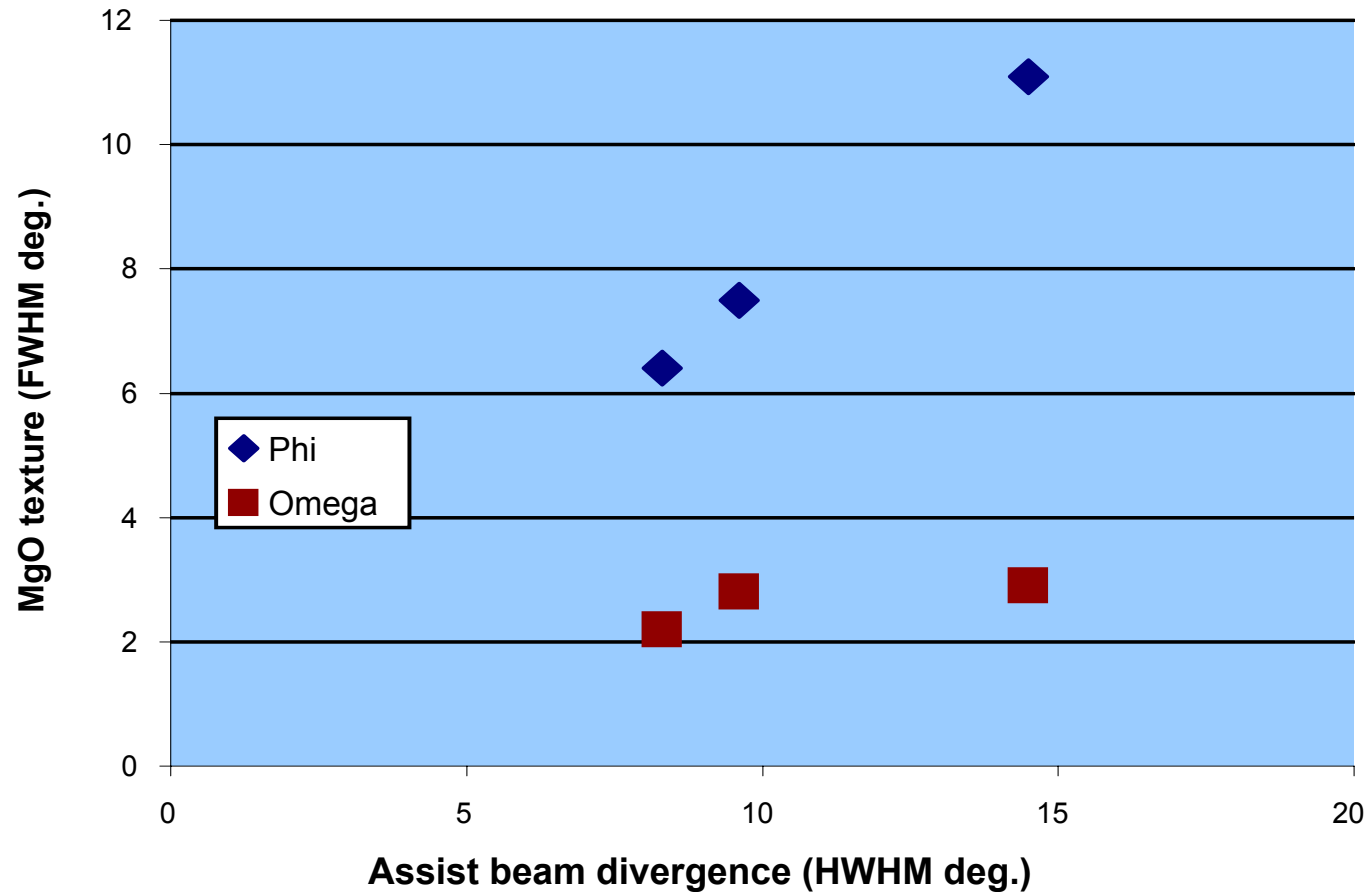


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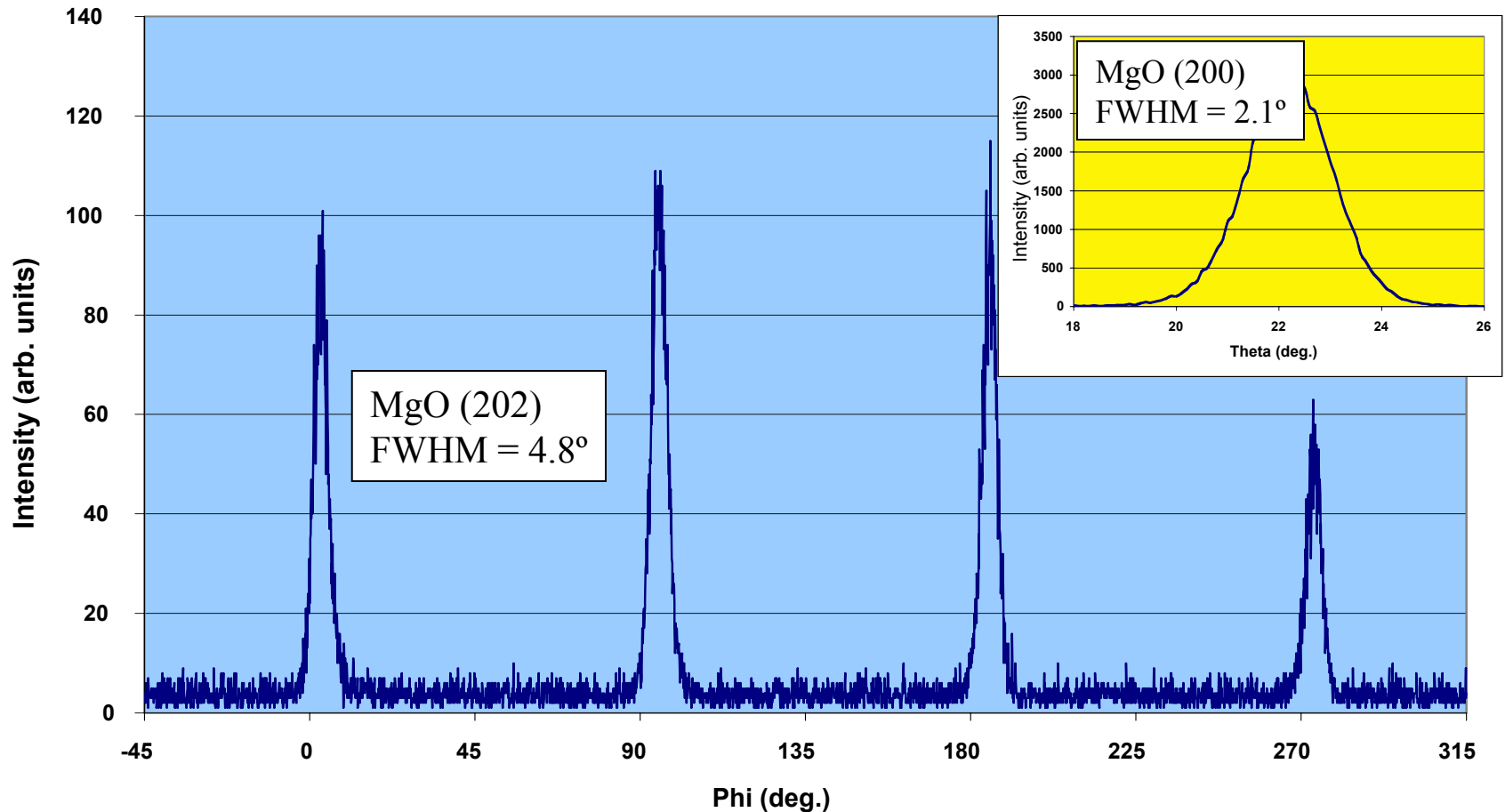
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IBAD MgO texture is improved with decreased assist ion beam divergence



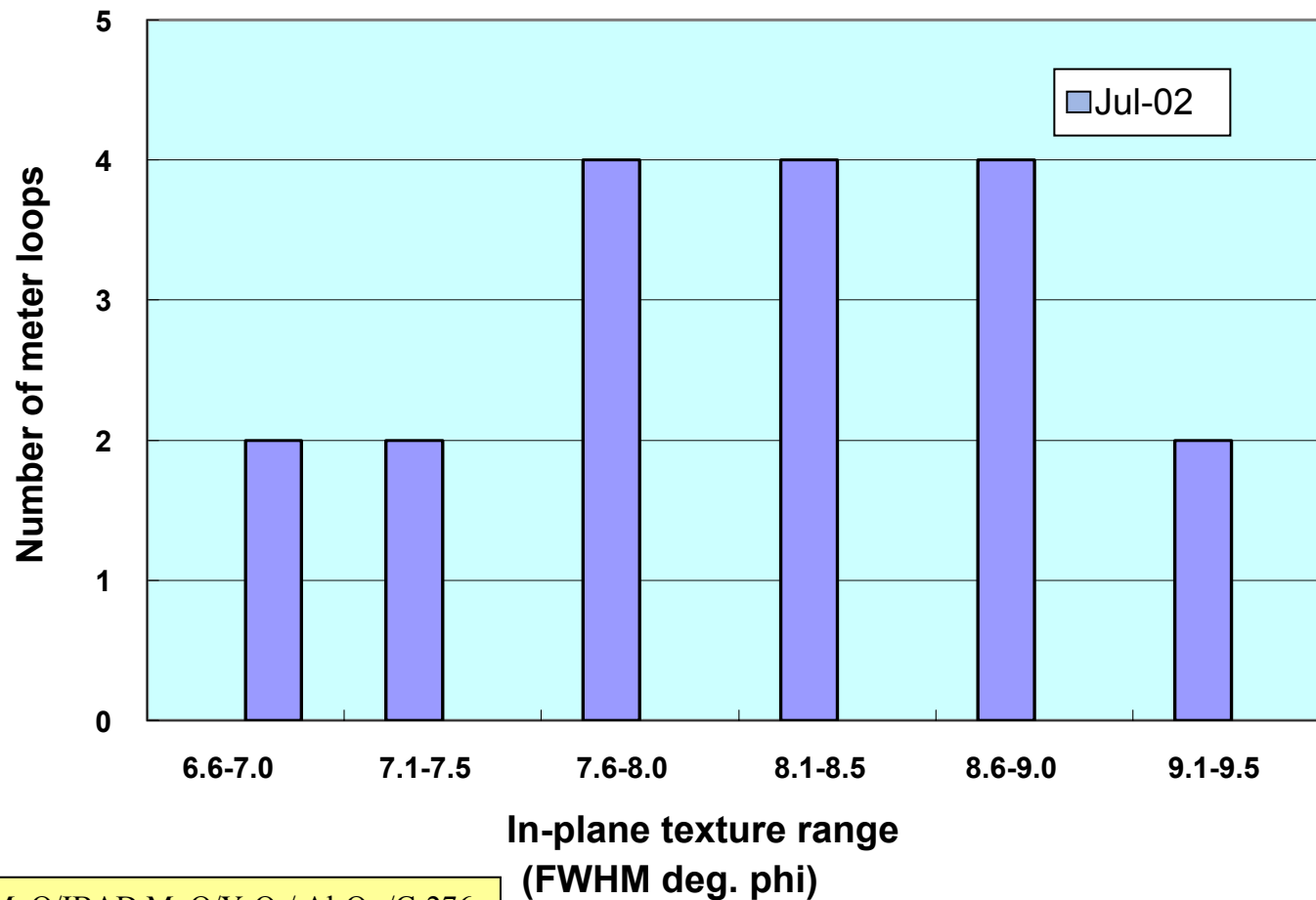
Homoepi MgO/IBAD MgO/Y₂O₃/Si

Assist beam optimization experiments led to improved texture for IBAD MgO templates



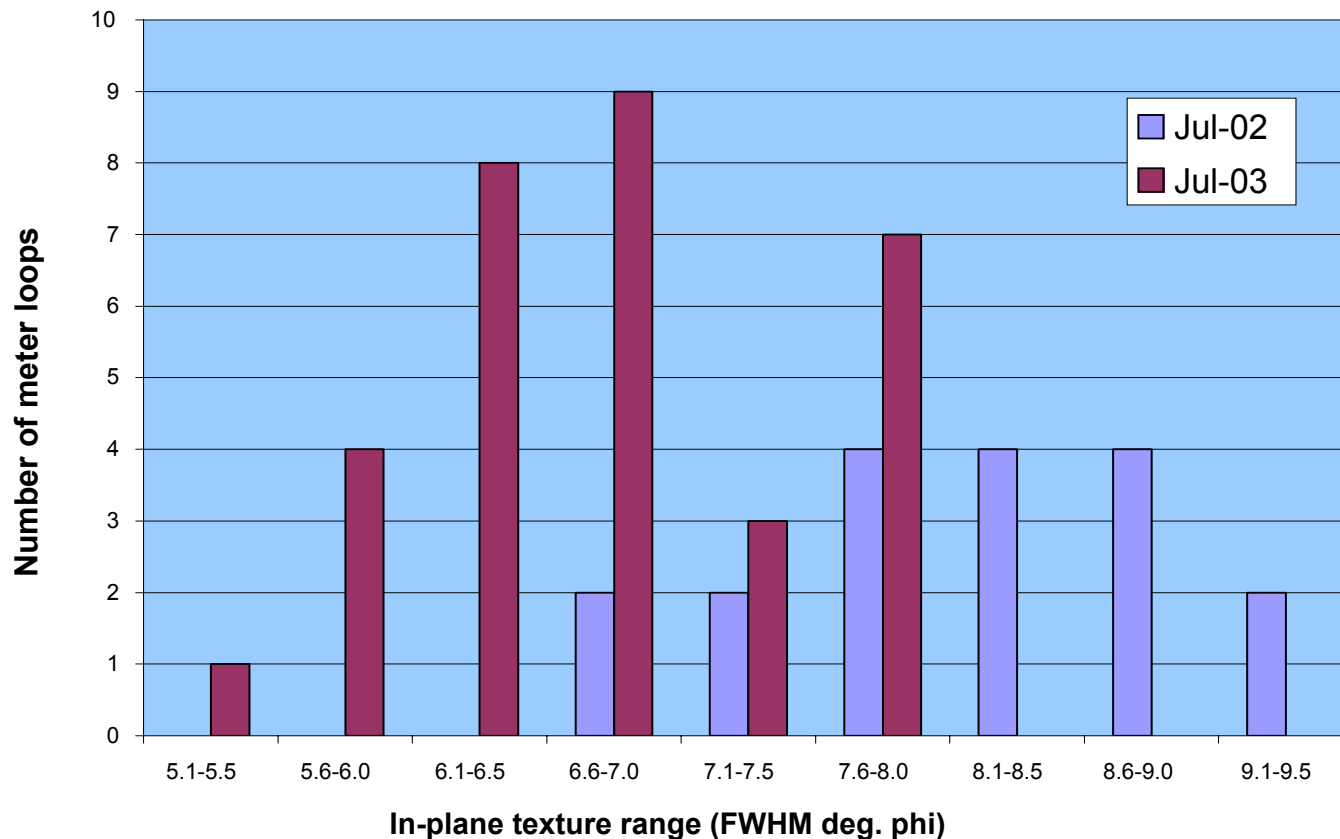
Homoepi MgO/IBAD MgO/Y₂O₃/Si

IBAD MgO in-plane texture - reproducibility of continuously processed meter loops



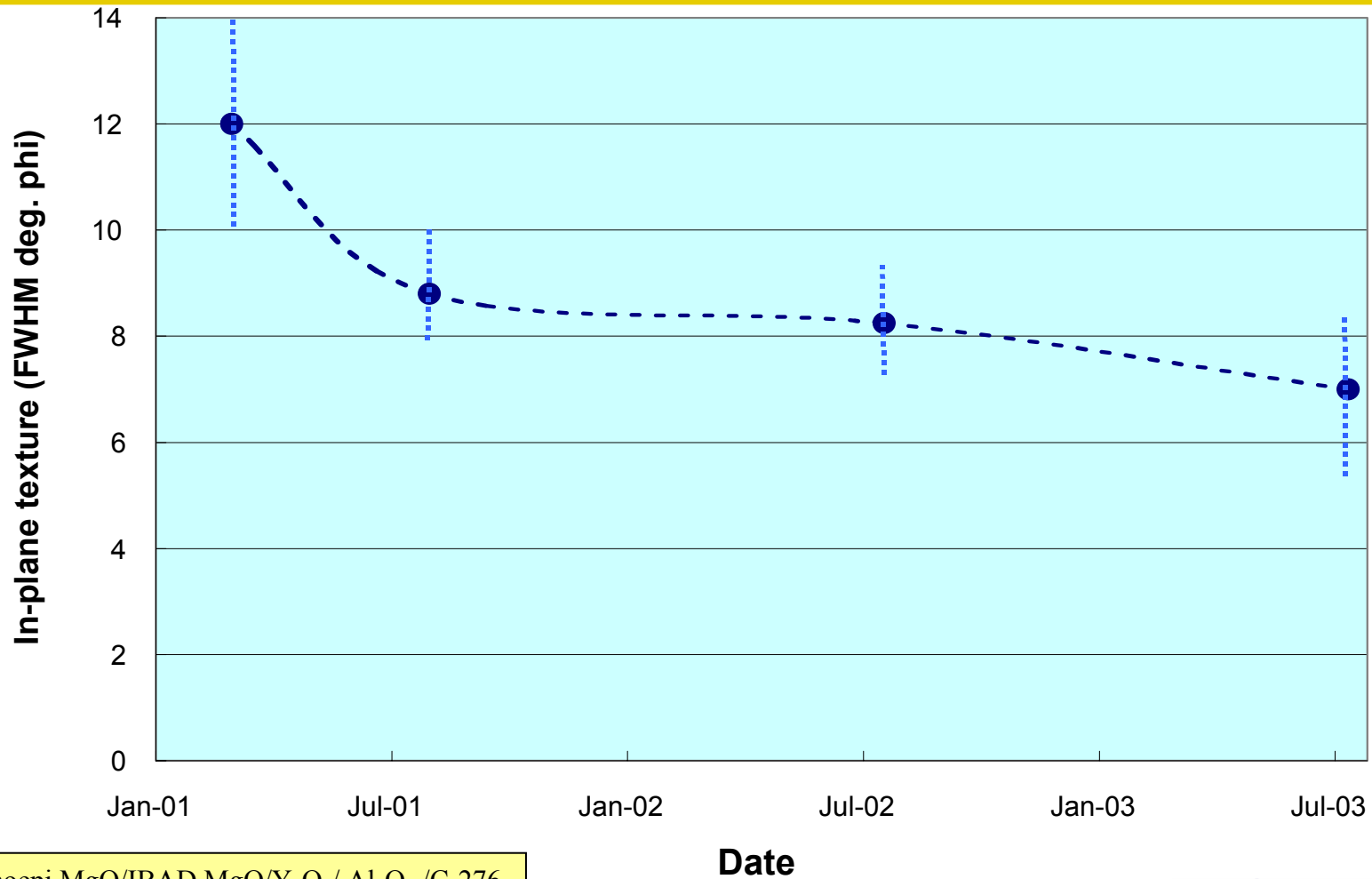
Homoepi MgO/IBAD MgO/Y₂O₃/ Al₂O₃ /C-276

IBAD MgO in-plane texture - reproducibility of continuously processed meter loops



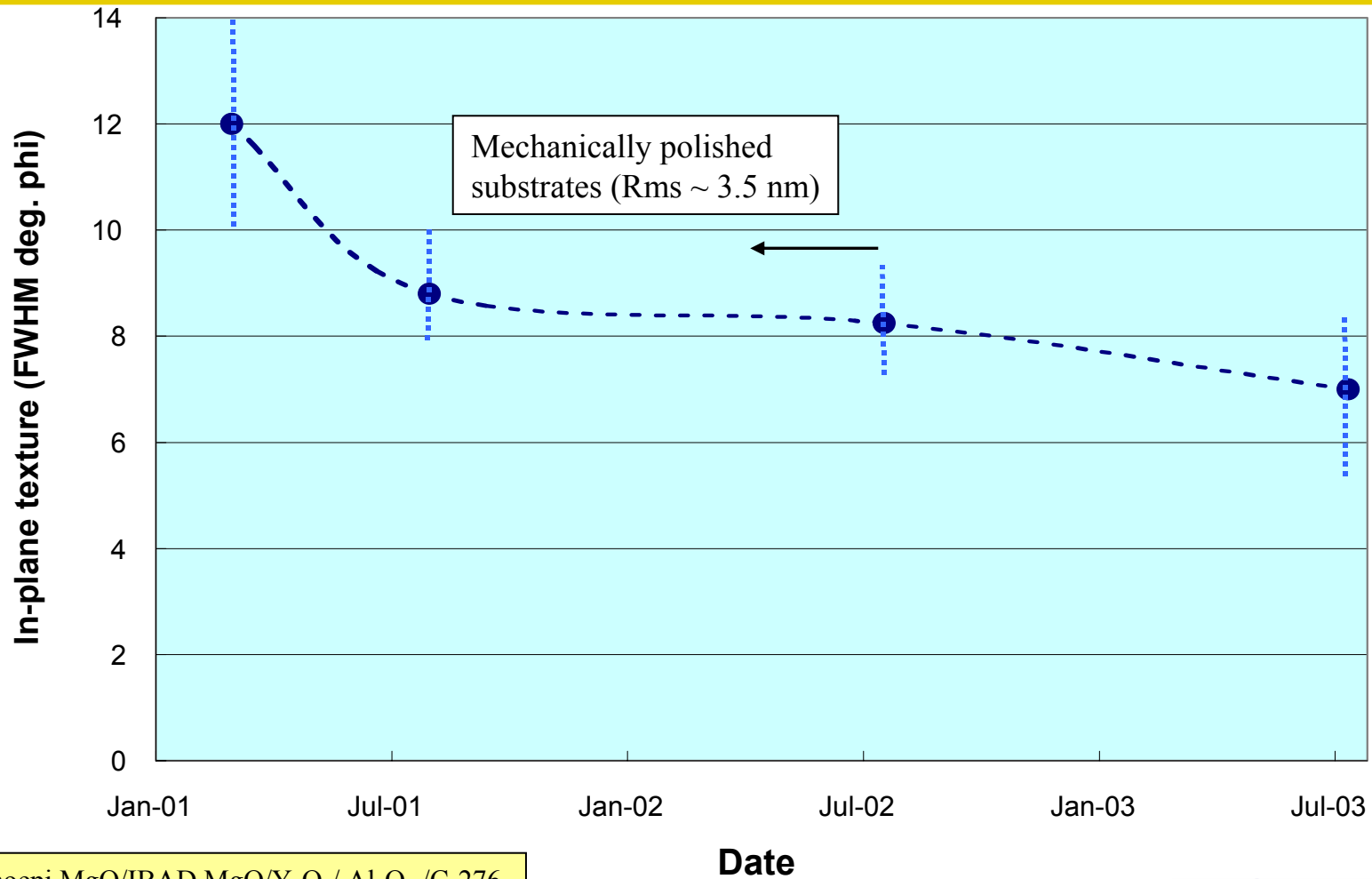
Homoepi MgO/IBAD MgO/Y₂O₃/ Al₂O₃ /C-276

Texture history for continuously processed IBAD MgO templates



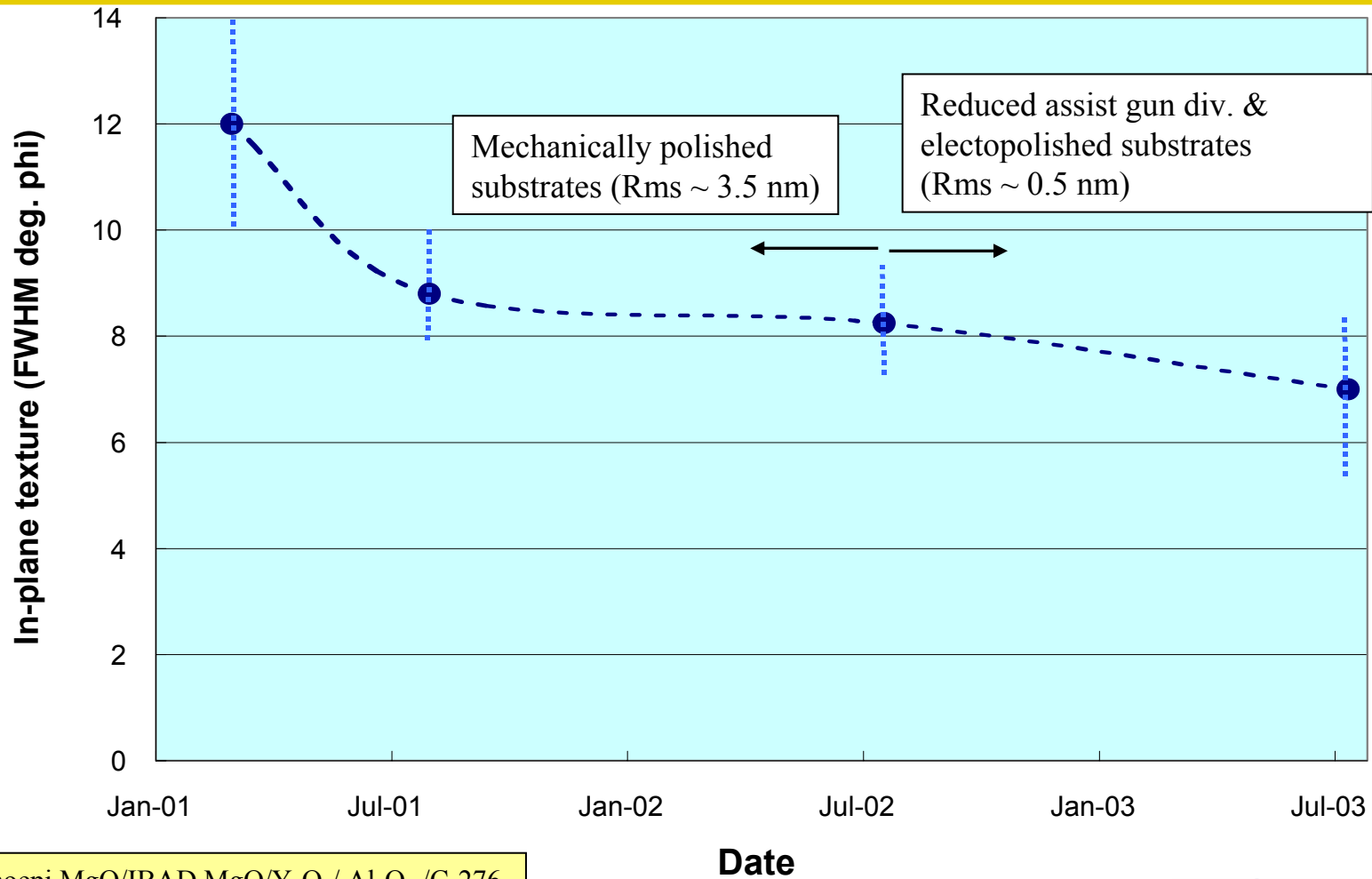
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Texture history for continuously processed IBAD MgO templates



Homoepi MgO/IBAD MgO/Y₂O₃/ Al₂O₃ /C-276

Summary

- Alumina inhibits diffusion of substrate cations to the YBCO layer
- Yttria extends the thickness window for IBAD MgO processing
- Erbium combines the barrier and nucleation properties of the alumina and yttria layers
- Reduction of the assist ion gun divergence leads to improved IBAD MgO texture
- Average in-plane texture of continuously processed meters has improved since FY02 by 1.5°

High current YBCO on IBAD MgO by pulsed laser deposition

Steve Foltyn, Paul Arendt, Leonardo Civale, Yates Coulter, Ray DePaula, Paul Dowden,
Randy Groves, Quanxi Jia, Yuan Li, Boris Maiorov, Liliana Stan, Haiyan Wang

*Superconductivity Technology Center
Los Alamos National Laboratory*

Judith MacManus-Driscoll (visiting staff member) – *University of Cambridge*

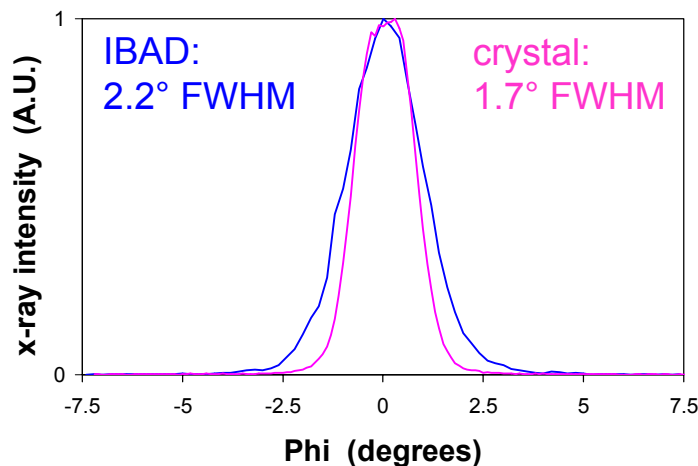
Jodi Reeves – *SuperPower, Inc.*

1. YBCO on IBAD MgO with near-single-crystal texture
2. High critical current YBCO on IBAD MgO
3. High I_c continuously processed tapes

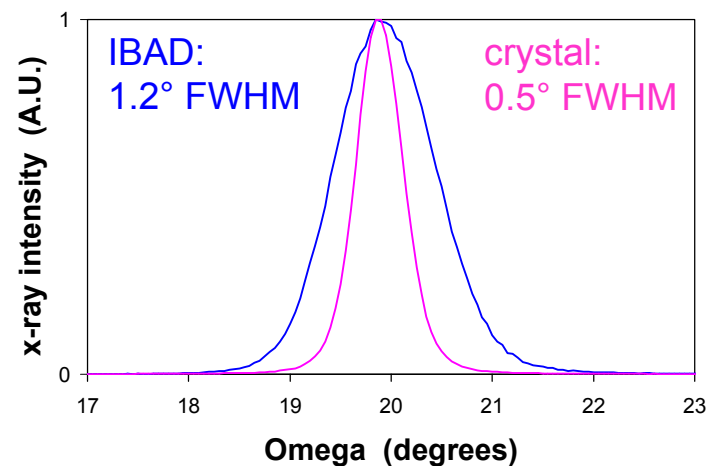
Improved alignment in IBAD MgO has enabled the achievement of near-single-crystal texture for YBCO

With IBAD MgO texture in the 7° FWHM range, YBCO texture is approaching that measured on single crystal MgO (YBCO thickness: $\sim 1.8 \mu\text{m}$)

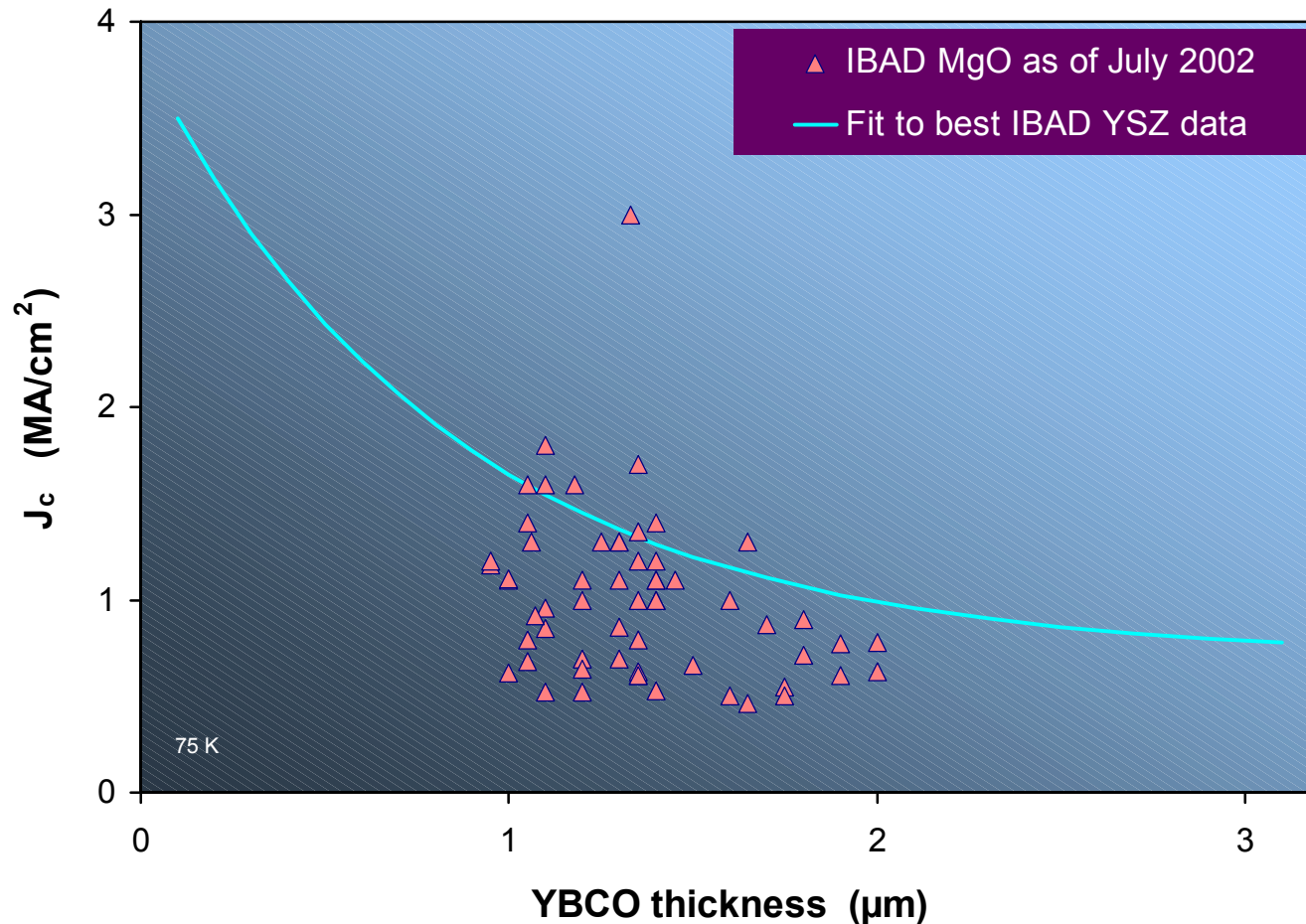
Phi-scan on YBCO (103)



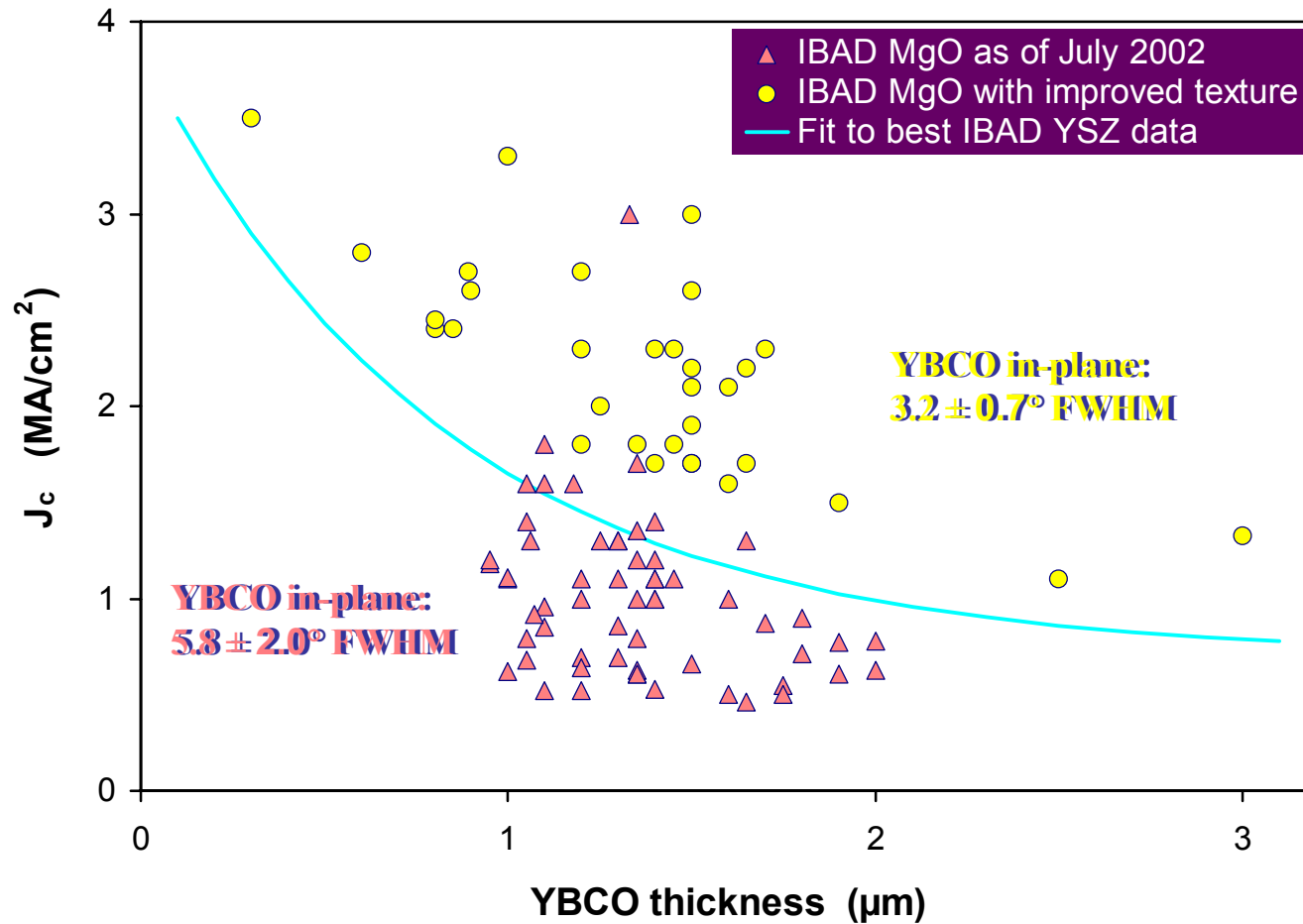
Rocking curve on YBCO (005)



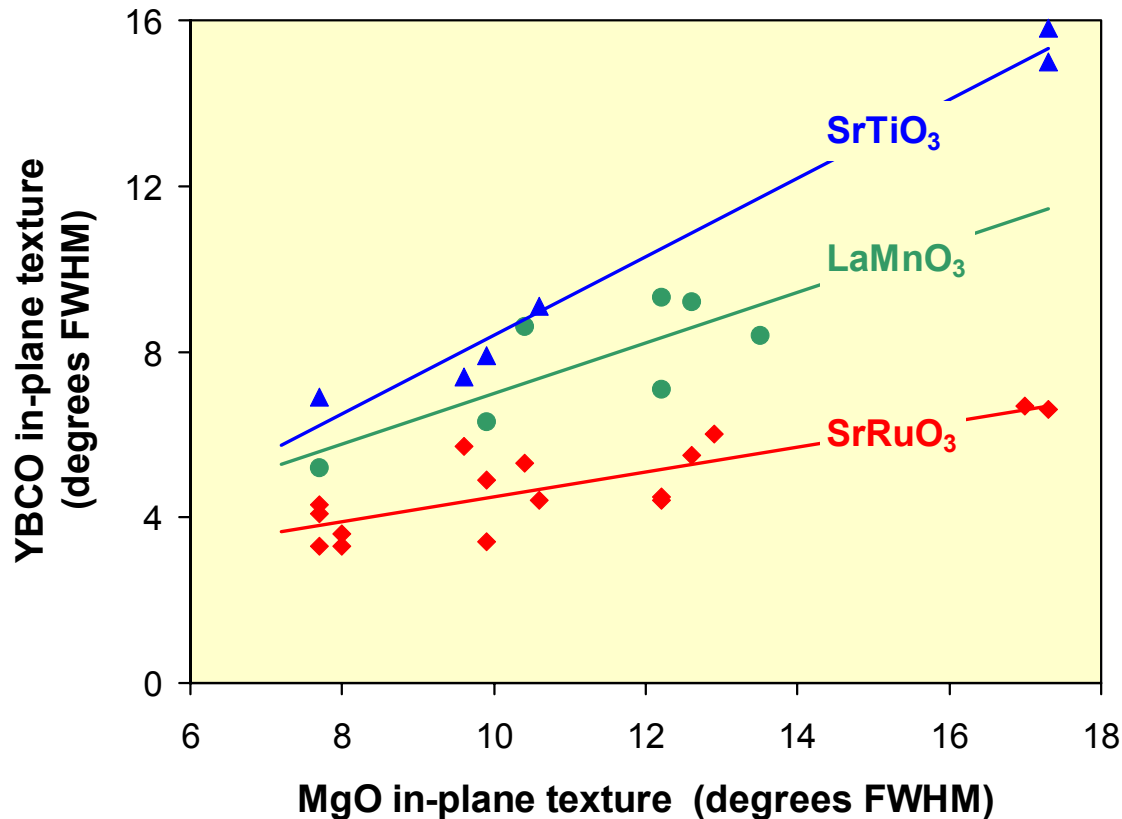
Last year, our J_c levels for IBAD MgO samples were generally below those for IBAD YSZ ...



... but improved in-plane texture now routinely yields higher performance



Part of the performance increase resulted from switching the buffer material from SrRuO_3 to SrTiO_3

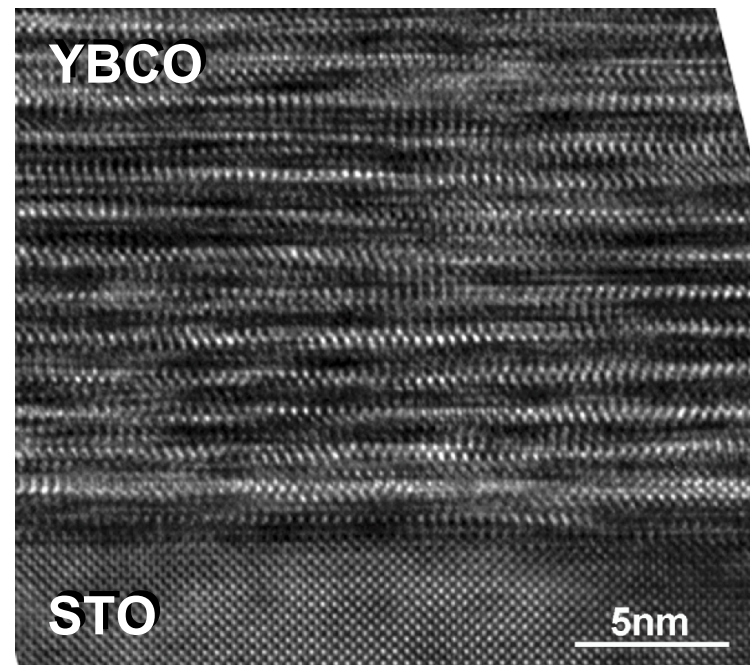
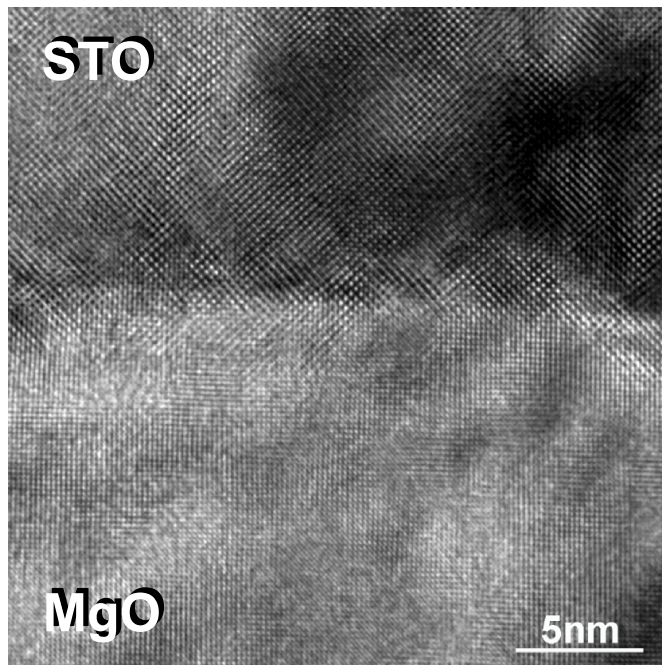


Because SrTiO_3 does not improve YBCO texture as much as LaMnO_3 or SrRuO_3 , its use is not feasible unless the IBAD MgO texture is less than $\sim 8^\circ$ FWHM.

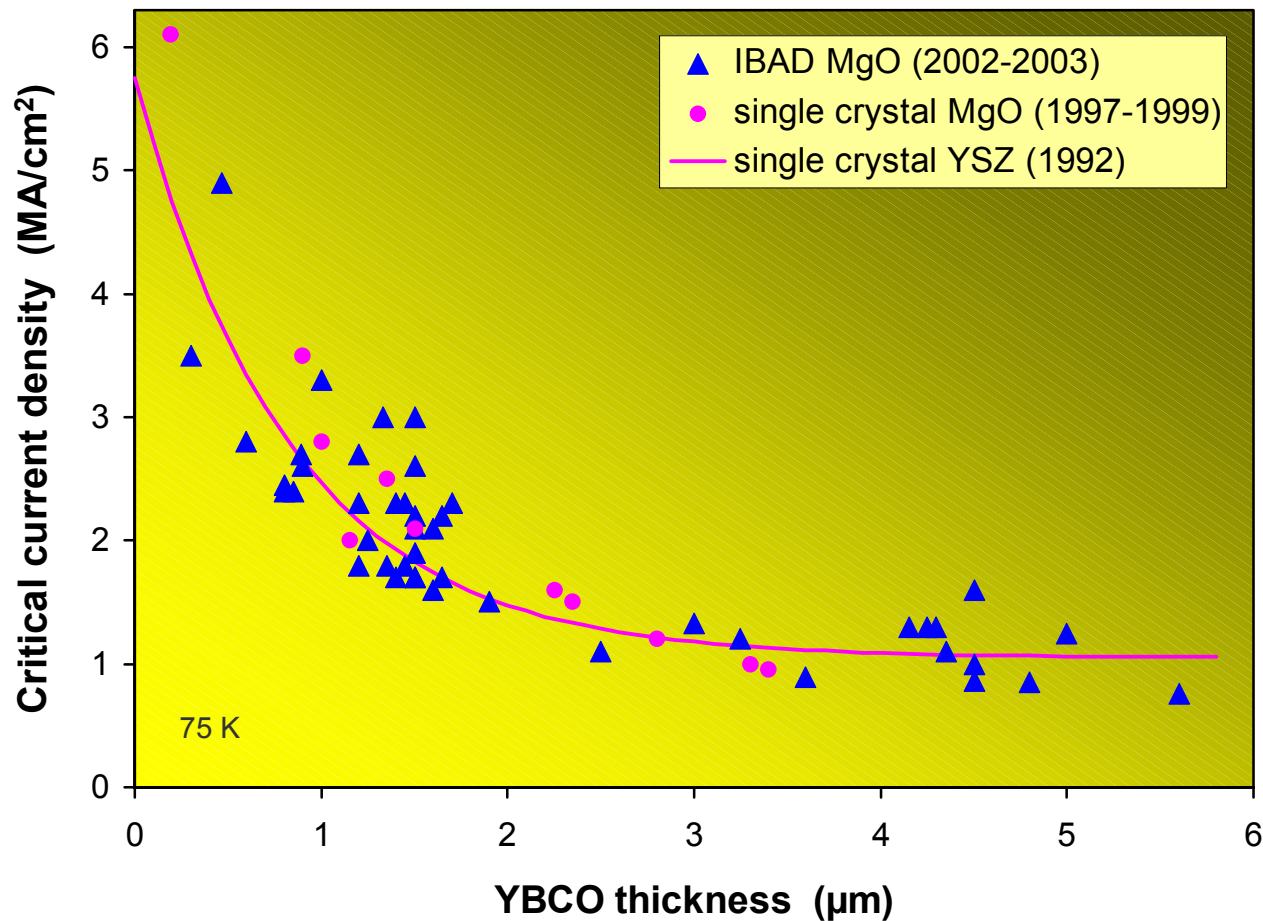
Data presented at 2002 Peer Review

SrTiO_3 is a chemically stable buffer material that readily aligns on MgO and is an excellent lattice match for YBCO

High-resolution TEM cross-sections from an IBAD MgO sample reveal that both the top and bottom interfaces of the SrTiO_3 buffer layer are smooth and free of reaction products.

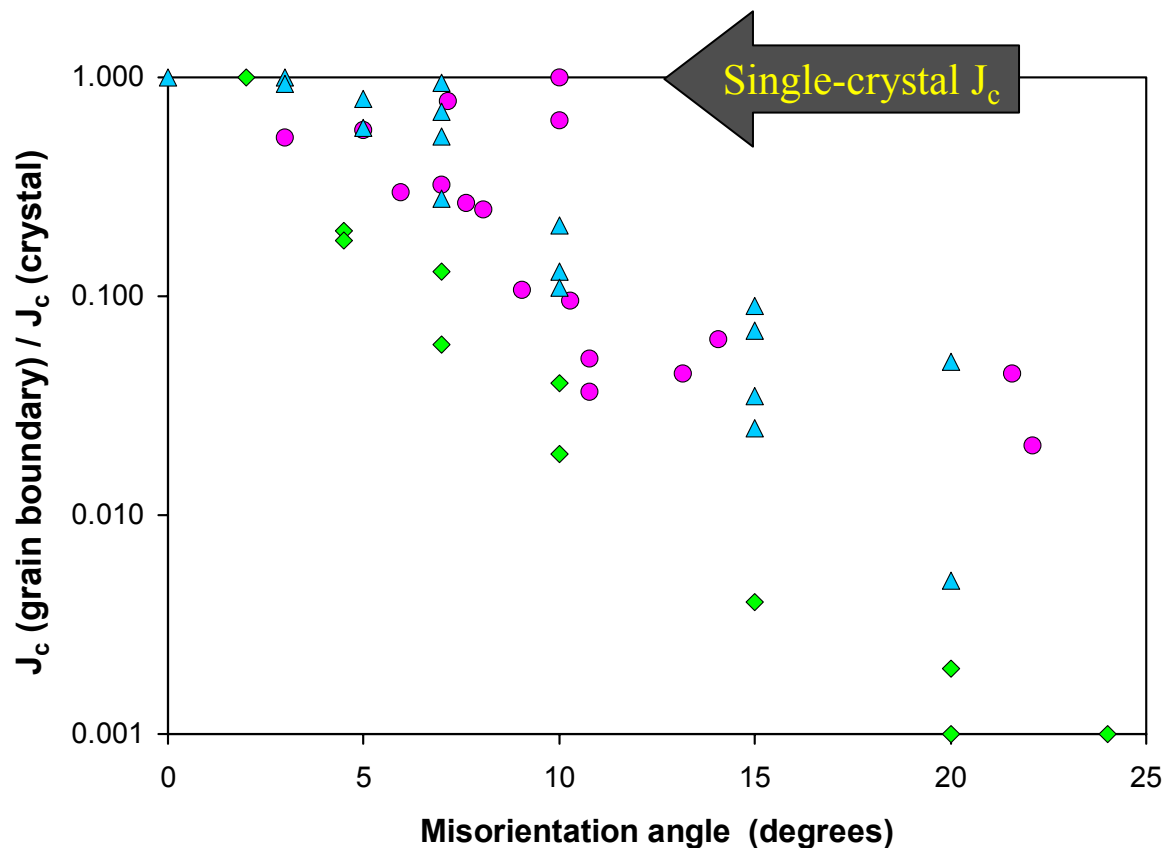


Improved texture yields IBAD MgO J_c s that are comparable to those for single-crystal substrates



One argument is that the often-observed bicrystal plateau occurs in coated conductors, too

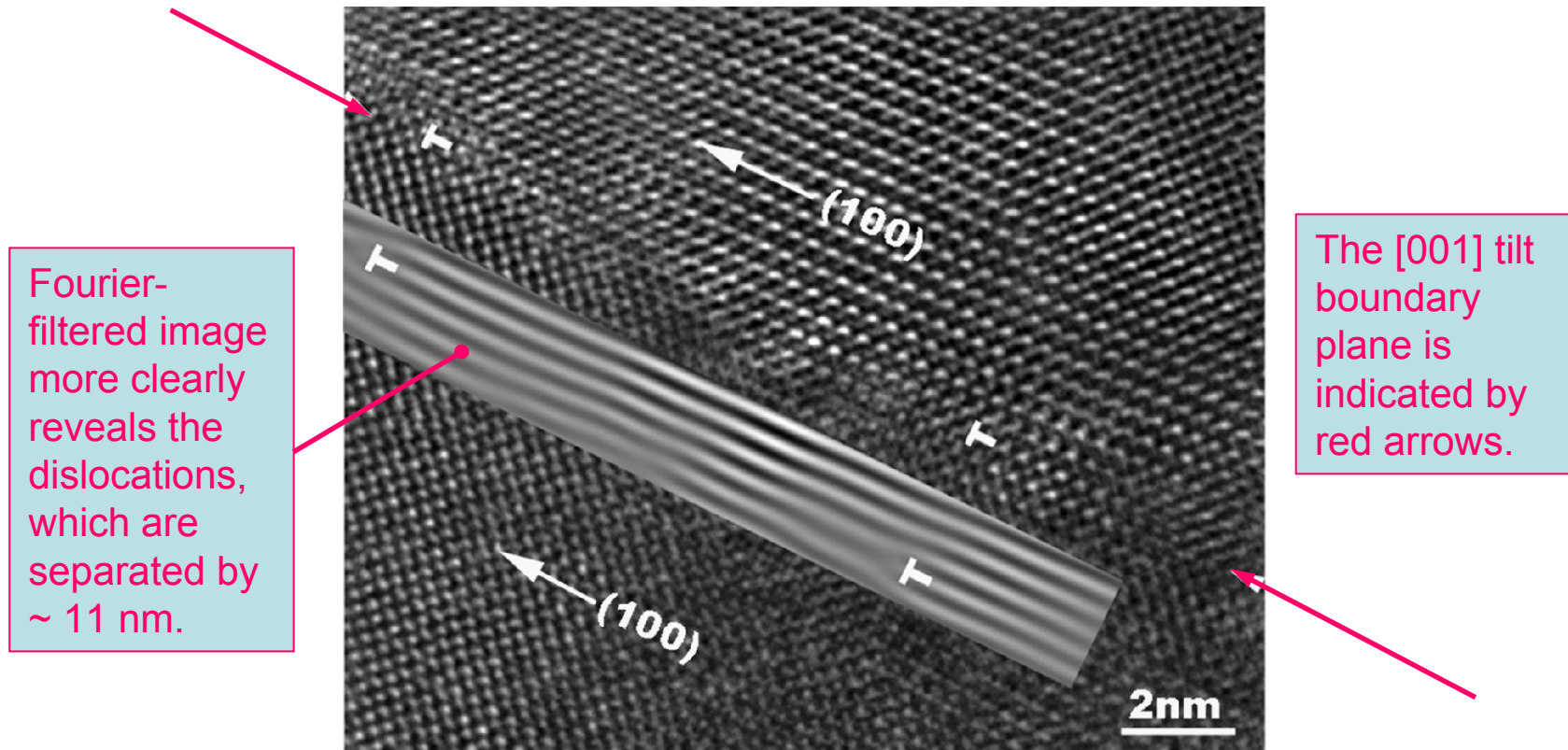
In bicrystals, perfect alignment is not needed to reach single crystal J_c levels



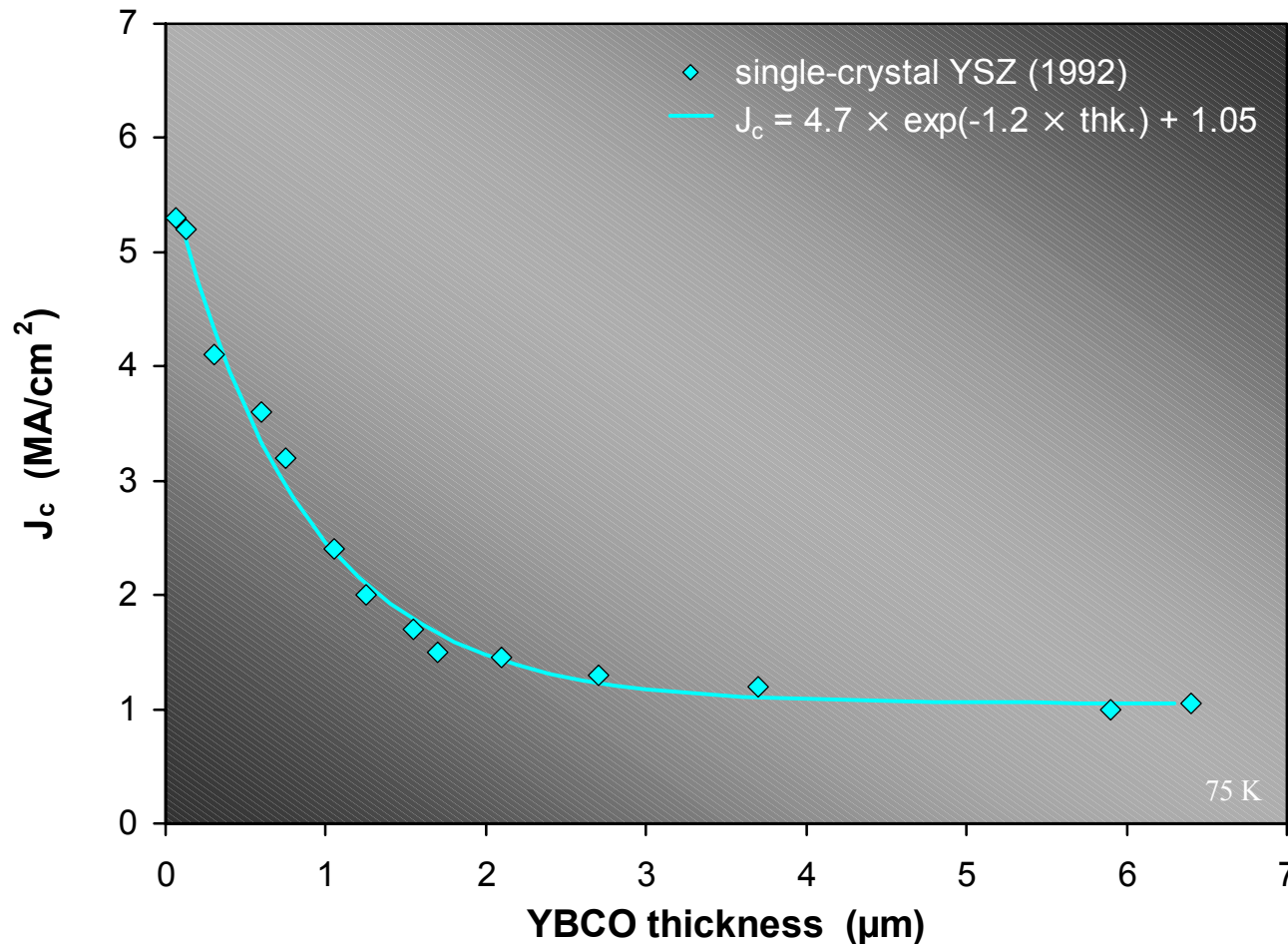
- D. Dimos, *et al.*, Phys. Rev. B **41**, 4038 (1990).
- ▲ N.F. Heinig, *et al.*, Appl. Phys. Lett. **69**, 577 (1996).
- ◆ D. T. Verebelyi, *et al.*, Appl. Phys. Lett. **76**, 1755 (2000).

YBCO crystallinity is barely disrupted in very low angle grain boundaries

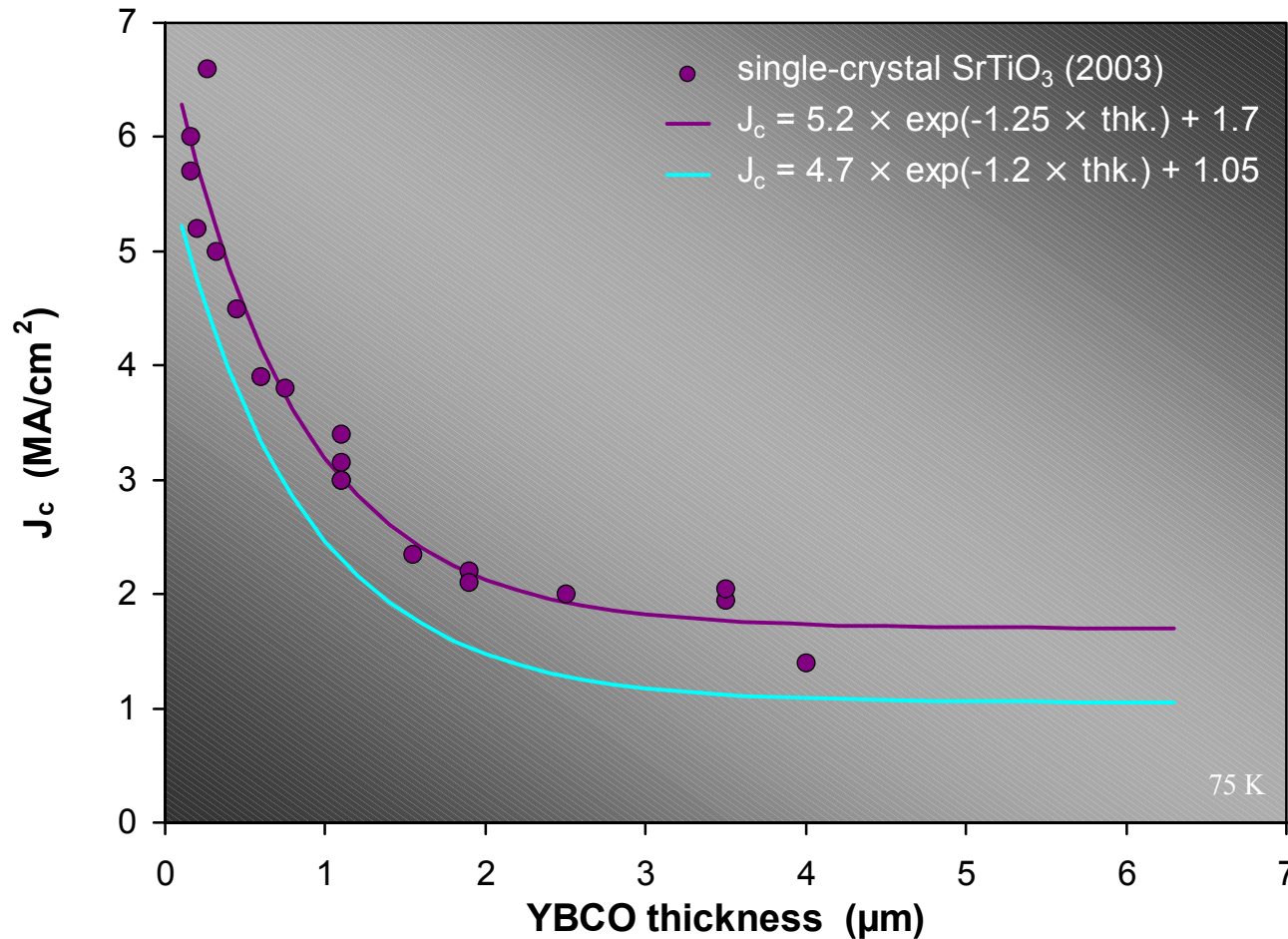
Plan-view TEM showing 2.2 degree grain boundary in YBCO on IBAD MgO



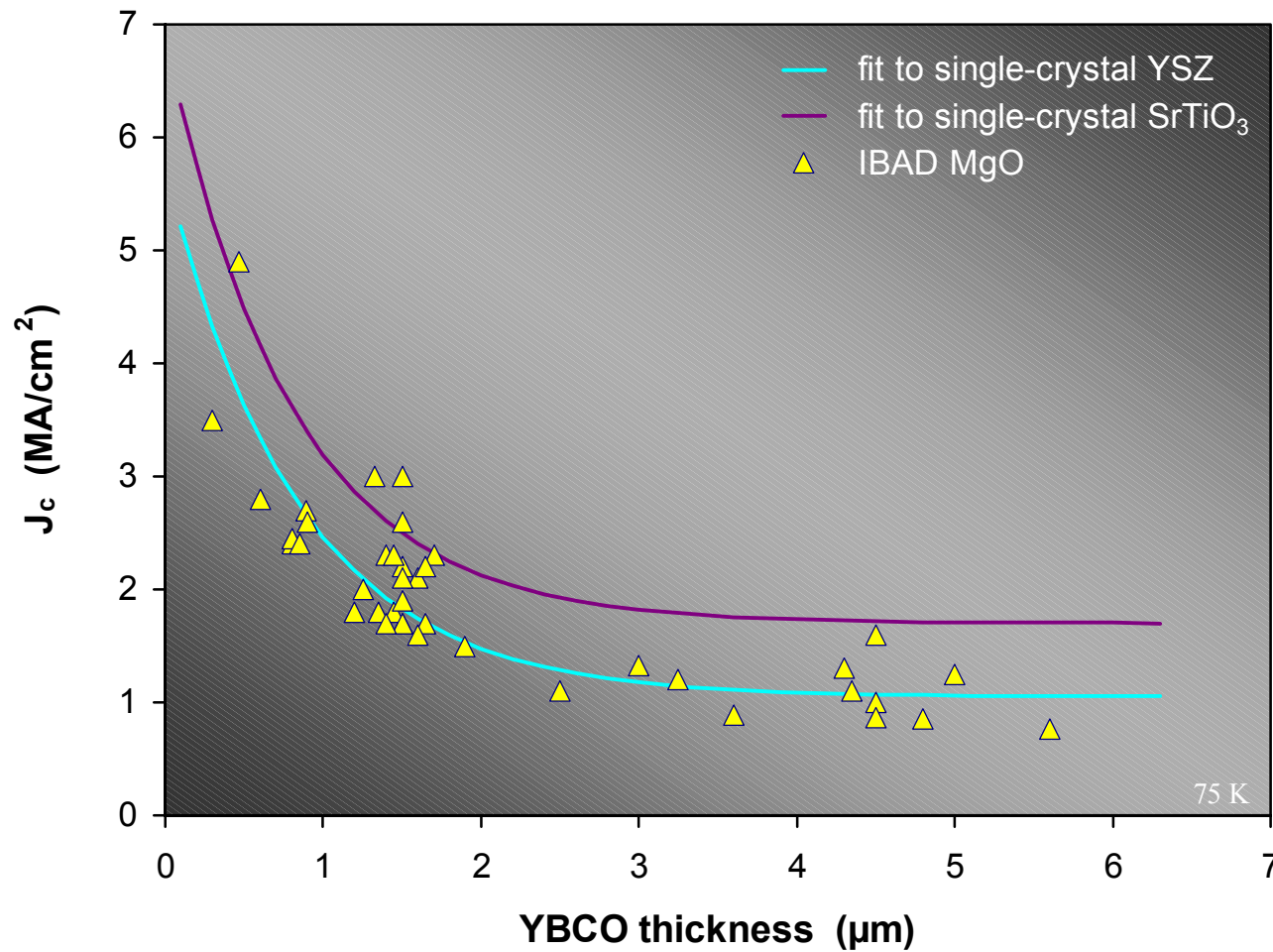
Another argument is that the 1992 single-crystal curve is an outdated benchmark ...



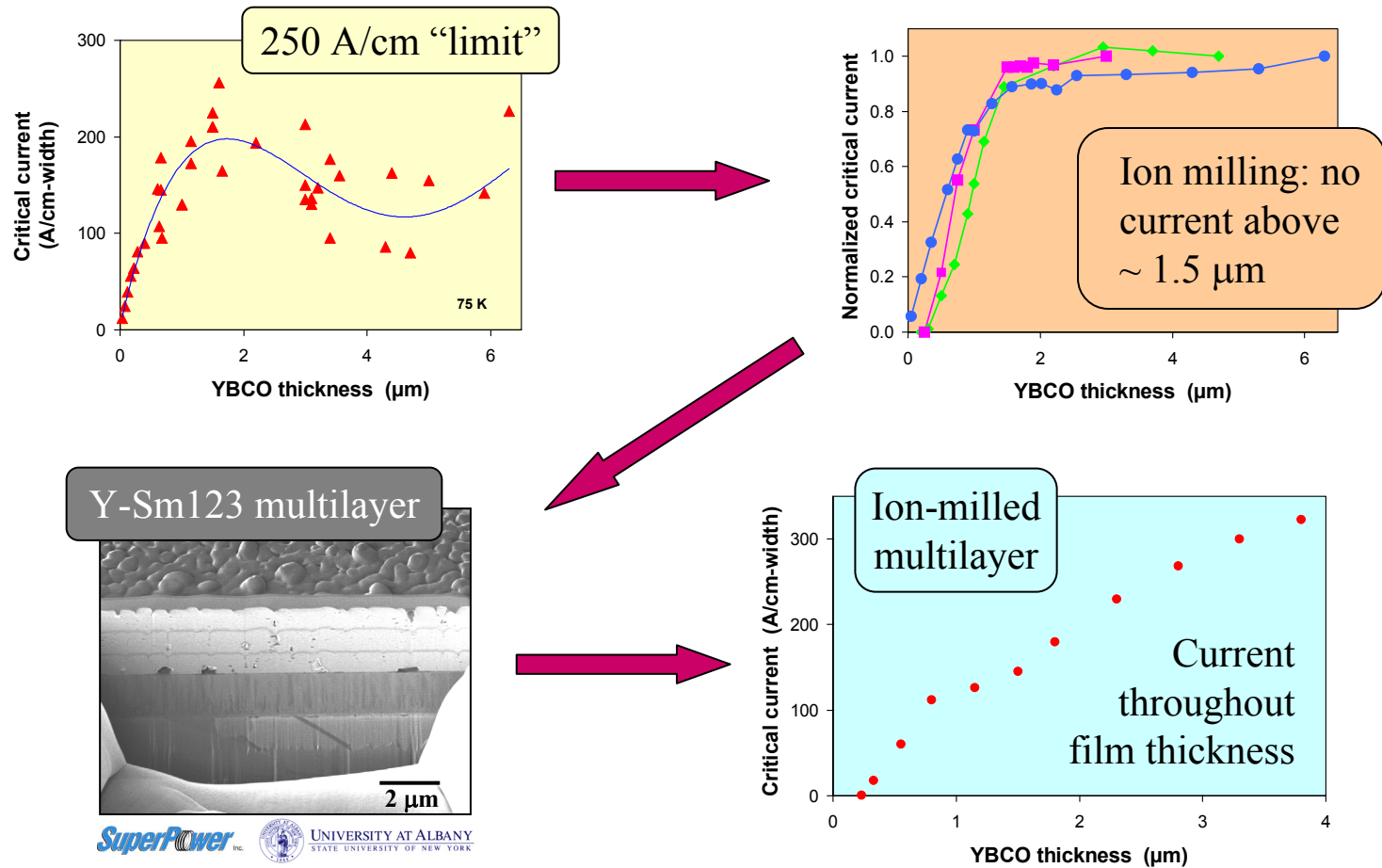
... so a more modern data set was obtained using the best possible substrate: single-crystal SrTiO_3



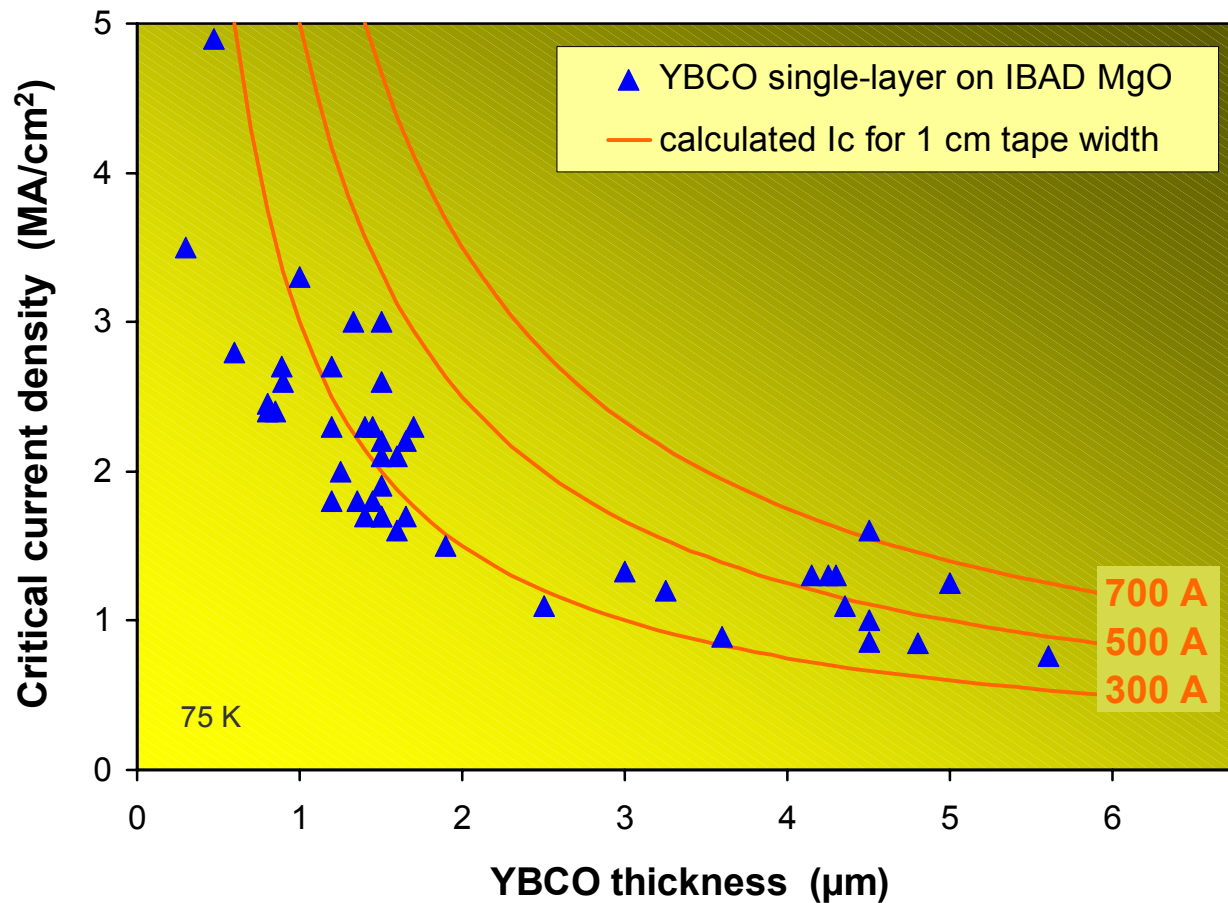
And the highest IBAD MgO J_c s still compare favorably with this new benchmark



We previously reported that we needed multilayers to reach I_c s over ~ 250 A/cm-width on IBAD YSZ;

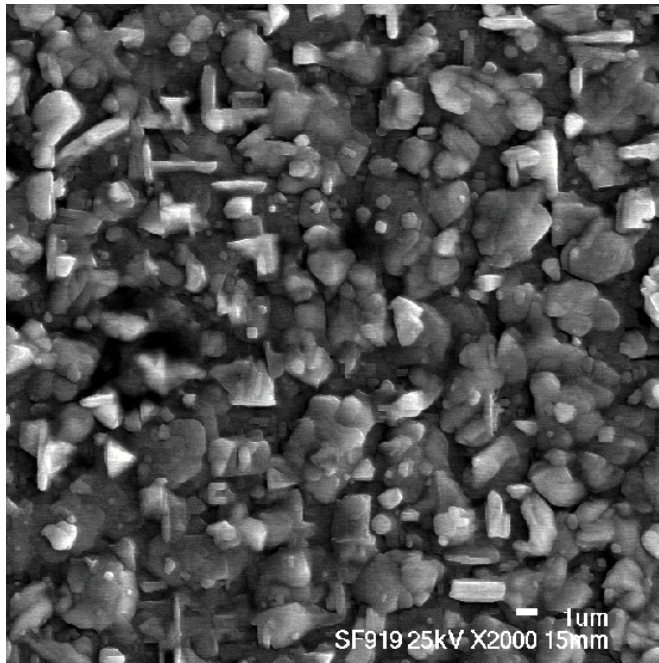


however, with IBAD MgO, multilayers are not needed to reach high current levels

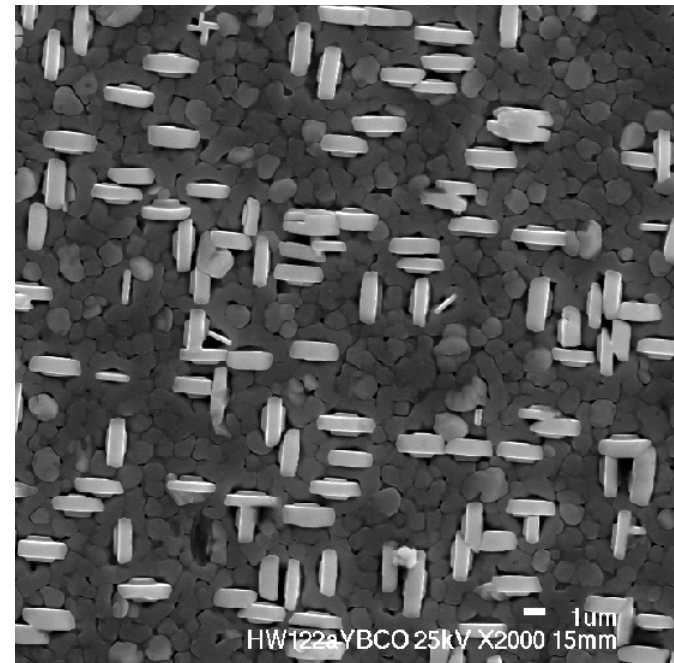


The difference is likely due to the smoother substrate, which reduces cumulative roughening of the YBCO

3.0 μm YBCO on IBAD YSZ
 $J_c = 0.6 \text{ MA/cm}^2$

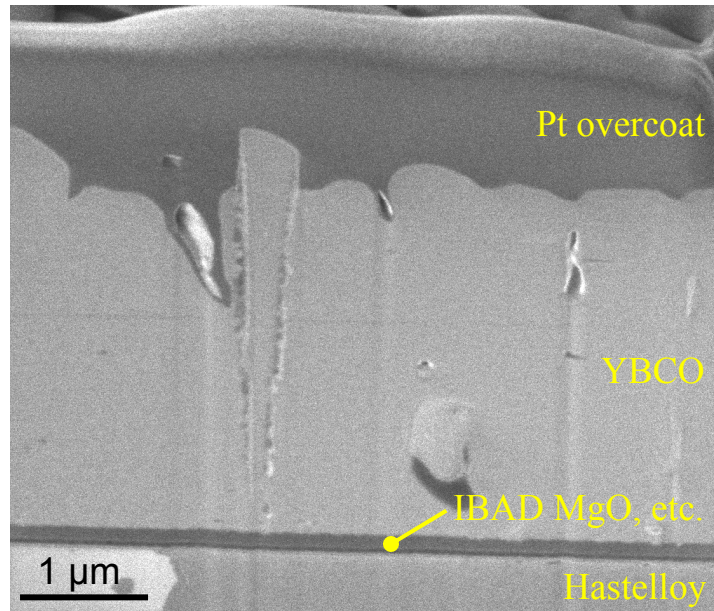


4.5 μm YBCO on IBAD MgO
 $J_c = 1.6 \text{ MA/cm}^2$



Focused ion beam cross-section shows a dense microstructure, largely free of porosity

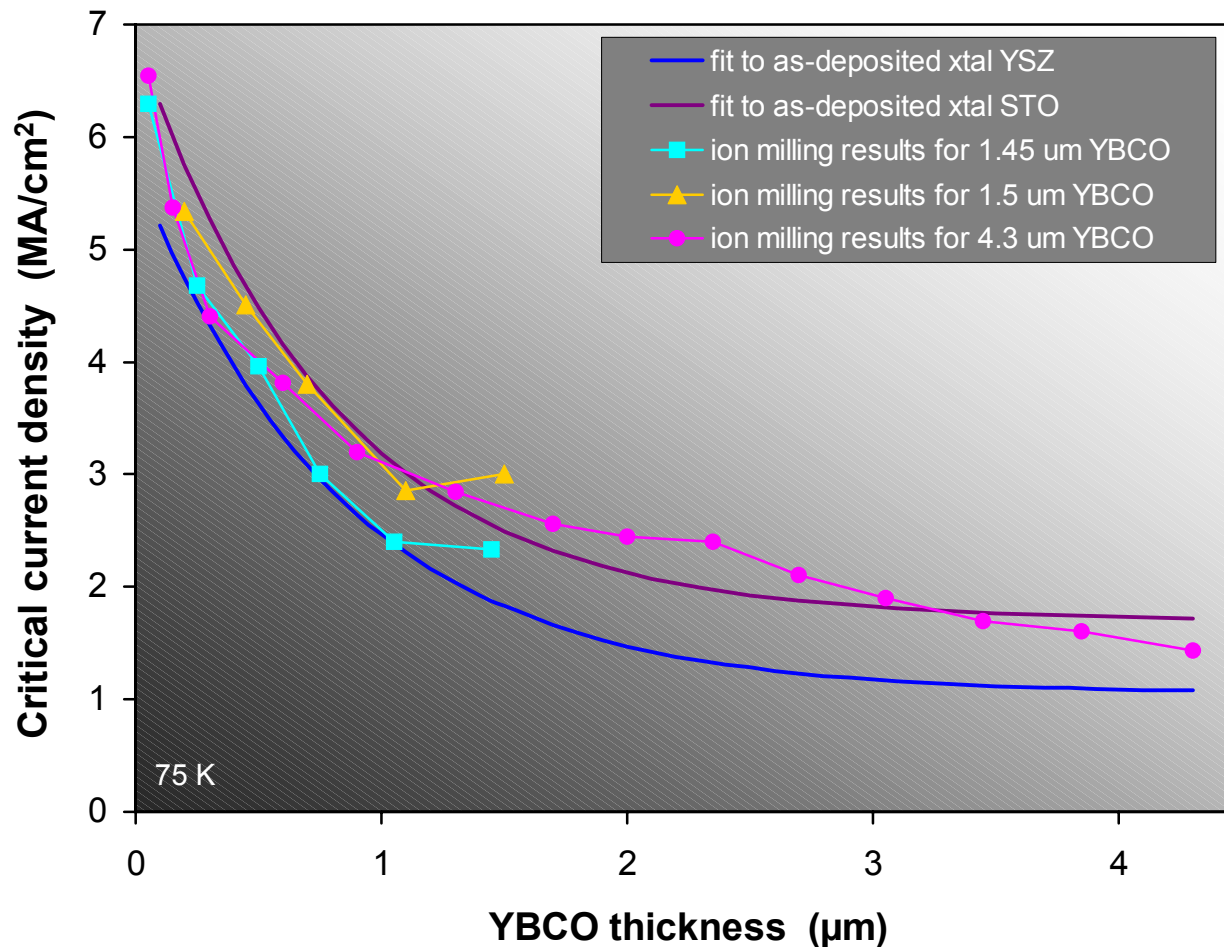
Cross-section (viewed at 45°) of a 4.5 μm YBCO film with $J_c > 1.0 \text{ MA/cm}^2$



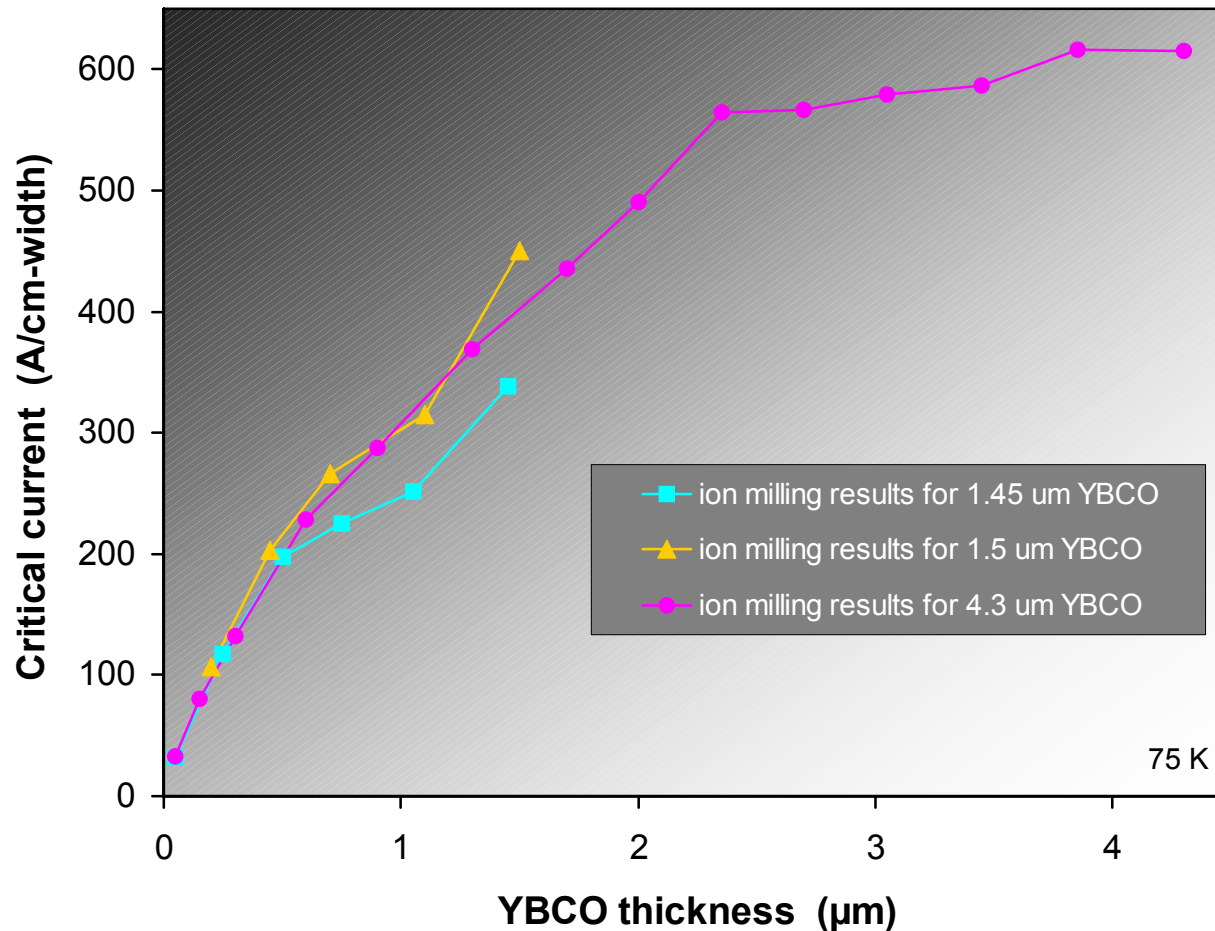
SuperPower Inc.

UNIVERSITY AT ALBANY
STATE UNIVERSITY OF NEW YORK

Ion milling shows that the dense structure allows J_c to remain high throughout the film thickness ...



... and allows I_c to continue increasing well beyond 1.5 microns of film thickness



Scoring criterion: FY 2003 Performance

“Optimize the thickness of each layer in the IBAD MgO architecture with a goal of reducing the total nonsuperconducting layer thickness to below 100 nm without reducing performance.”

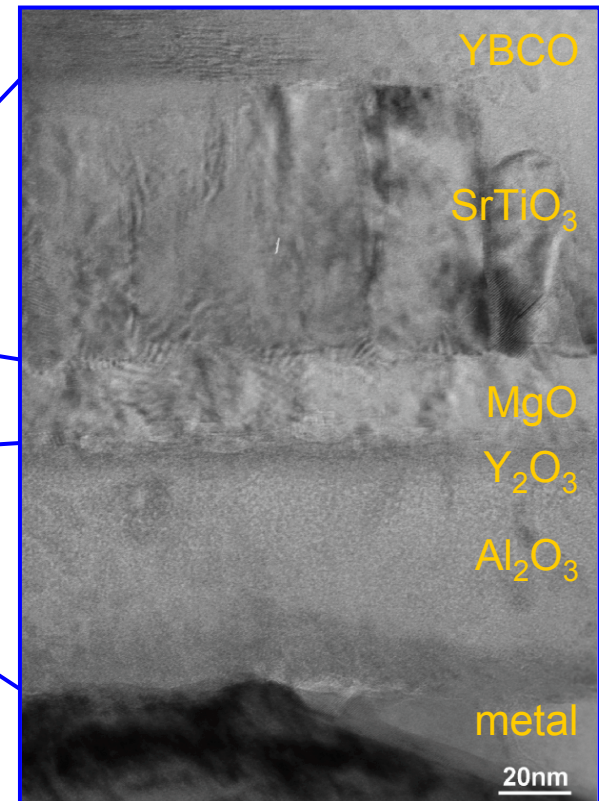
- Determined that $T_c < 75$ K unless the Al_2O_3 barrier layer is used – initially concluded that the Al_2O_3 cannot be eliminated.
- SIMS analysis revealed that this layer greatly reduces – but does not eliminate – diffusion of substrate elements into the YBCO, so we were reluctant to reduce barrier layer thickness.
- Recent ion milling results indicate that a certain amount of diffusion can be tolerated, so thickness reduction may be possible after all.
- Found that the Al_2O_3 barrier layer and the Y_2O_3 nucleation layer can be replaced by a single, 20nm layer of Er_2O_3 for a total nonsuperconducting layer thickness of 92 nm. J_c : 2.3 MA/cm² @ 1.7 μm .

Scoring criterion: FY 2003 Performance (continued)

- However, the Er_2O_3 layer substitution has thus far produced mixed results; for now, our preferred architecture is:

<u>YBCO</u>	<u>1-5 μm</u>
<u>SrTiO_3</u>	<u>70 nm</u>
<u>homoepitaxial MgO</u>	<u>13 nm</u>
<u>IBAD MgO</u>	<u>10 nm</u>
<u>Y_2O_3 nucleation layer</u>	<u>7 nm</u>
<u>Al_2O_3 barrier layer</u>	<u>55 nm</u>
<u>Hastelloy C-276 substrate</u>	<u>100 μm</u>

Total nonsuperconducting
layer thickness: 155 nm



Scoring criterion: FY 2003 Performance (continued)

“Support both IGC and the Research Park in scaling up the latest IBAD and PLD technologies.”

- Continued IBAD and PLD technology transfer to SuperPower via frequent communication, site visits, sample and data exchange, and licensing of intellectual property.
- SuperPower has made substantial progress this year, as outlined in their presentation on Wednesday.
- Supported the Research Park through regular interactions, technician support, and equipment loans; also collaborated on small-sample and tape test runs to confirm performance of Research Park IBAD MgO and PLD buffer layers.
- All substrate tape used in the Core Program was reel-to-reel electropolished by Research Park personnel.

Scoring criterion: FY 2003 Performance (continued)

“Distribute IBAD MgO to our industrial partners and national laboratory collaborators for deposition of YBCO, and work with them individually to achieve optimum performance with their respective deposition processes.”

- Sent samples of IBAD MgO with various buffer layers to American Superconductor, and assisted in the development of AMSC's CeO_2 cap layer.
- Also sent IBAD MgO samples to Oak Ridge for deposition of LaMnO_3 – these samples were then sent to AMSC for MOD YBCO.
- Best results for batch-processed samples (0.8-0.9 μm YBCO, 5 mm wide):
 - ❖ With $\text{SrRuO}_3/\text{CeO}_2$ – up to 1.8 MA/cm^2 (~ 140 A/cm-width)
 - ❖ With $\text{LaMnO}_3/\text{CeO}_2$ – up to 2.6 MA/cm^2 (~ 200 A/cm-width)

Scoring criterion: FY 2003 Performance (continued)

- Sent IBAD MgO tapes of various lengths to SuperPower, Inc., initially with a SrRuO_3 buffer layer, more recently with SrTiO_3 .
- Provided information about our PLD YBCO deposition conditions in order to facilitate optimization at SuperPower.
- Best results for 1-cm-wide continuously processed tape:
 - ❖ I_c values of 100 – 150 A on 3-5 cm lengths.
 - ❖ J_c of $1.5\text{MA}/\text{cm}^2$ on $0.9\text{ }\mu\text{m}$ thick YBCO
 - ❖ Data from seven runs with average I_c of $128\text{ A} \pm 14\text{ A}$

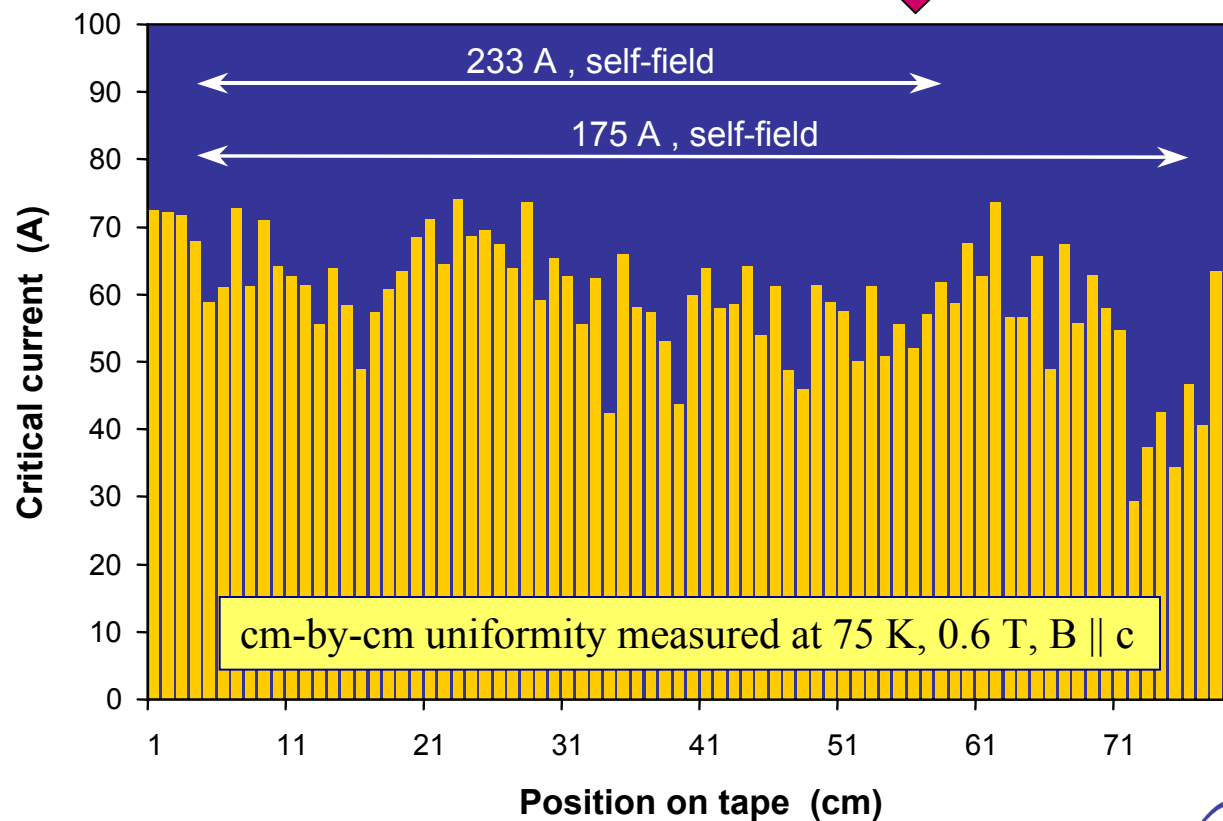
Scoring criterion: FY 2003 Performance (continued)

“Solve the problem(s) preventing us from achieving high-current YBCO on IBAD MgO/SrRuO₃ (Goal: 100 A in a meter) ...”

- As expected, the primary obstacle to high I_c with continuous processing was the PLD tape-coater, and *not* the IBAD MgO itself.
- The main difficulty was lack of reproducibility, which was traced to building cooling water temperature that exceeded laser specifications at irregular intervals.
- Once this was fixed (with a chilled water recirculator), optimization of laser deposition parameters proceeded normally.
- After optimization, five short tapes (~ 20 cm coated, ~ 10 cm measured) were produced. I_c s were 265, 276, 292, 298, and 319 A (75K, self field, one centimeter width).

Scoring criterion: FY 2003 Performance (continued)

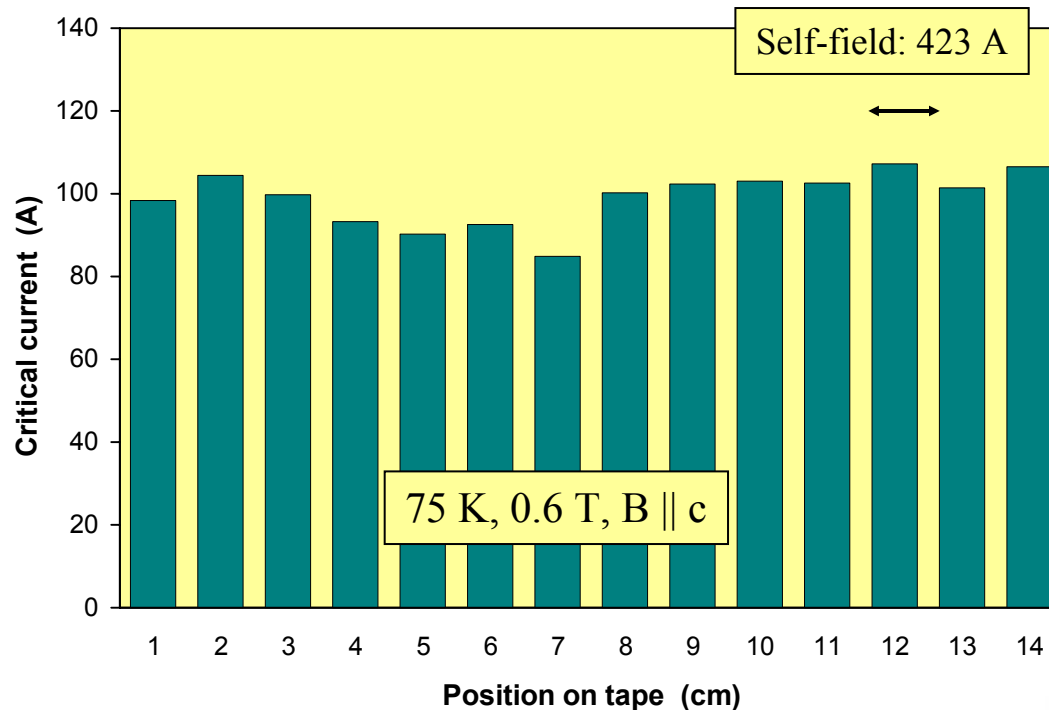
- Our first two longer lengths have self-field I_c s of 173 A over 70 cm, and 175 A over 72 cm (with 233 A over 54 cm).



Scoring criterion: FY 2003 Performance (continued)

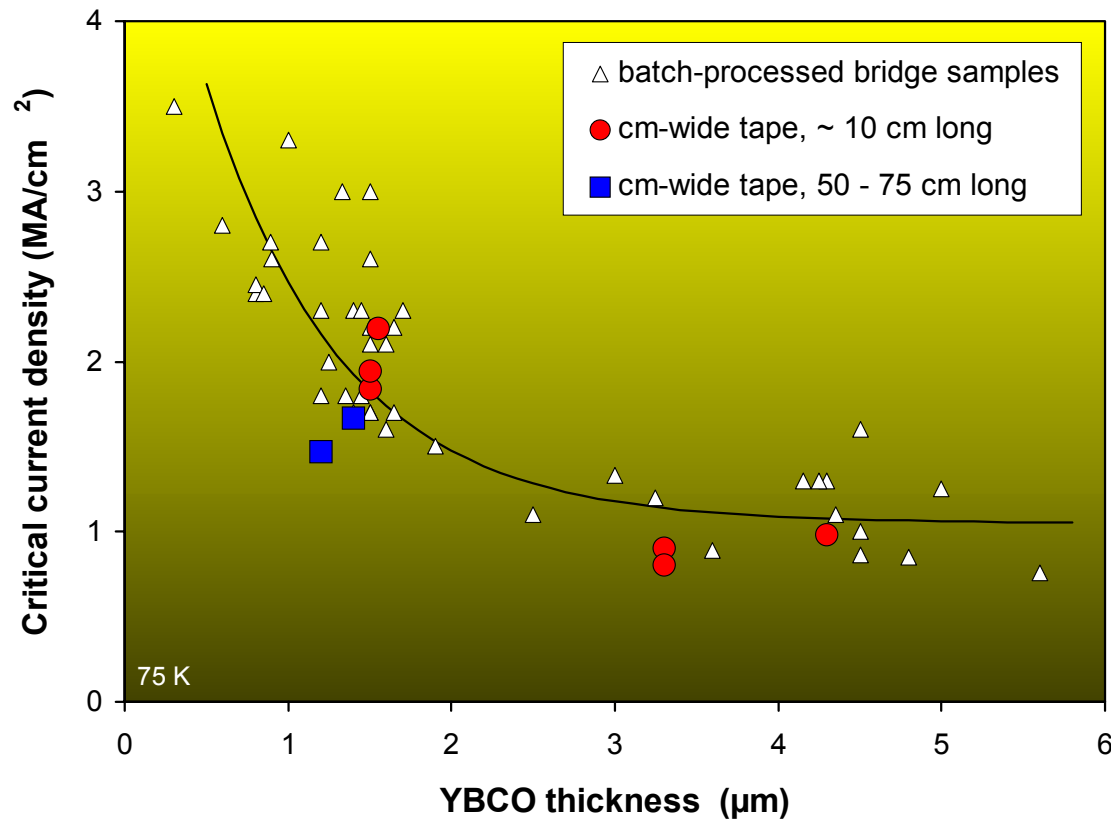
“...and incorporate multilayer technology to reach 200 A in an IBAD MgO meter.”

- Since multilayers are no longer necessary, we tried simply making the YBCO thicker ($4.3\text{ }\mu\text{m}$) with the following result:



Scoring criterion: FY 2003 Performance (continued)

- Continuously processed tape J_c s are comparable to batch-processed bridge results.



Scoring criterion: FY 2003 Performance (continued)

“Deposit YBCO on meter lengths of IBAD MgO with the Oak Ridge LaMnO₃ buffer layer. Goal: 100 A.”

- Based in part on good results obtained with Oak Ridge-sputtered LMO, the Research Park team has been routinely depositing LMO on their IBAD MgO, so we collaborated on this – using RP IBAD MgO/LMO – rather than duplicate the LMO effort in the Core Program.
- Several bridge samples were prepared with an average YBCO thickness of 1.3 μm , and an average J_c of 1.1 MA/cm² (140 A/cm-width). Best results were obtained with SrRuO₃ on top of the LaMnO₃.
- With SrTiO₃ on top of the Research Park’s IBAD/LMO, we reached an I_c of 251 A in a cm-wide x 4.5 cm long tape.

Scoring criterion: Research Integration

- * Distributed buffered IBAD MgO samples to SuperPower, Inc. and American Superconductor, and worked with each to assist in the deposition of high-quality YBCO on these samples. This is a successful first step in the transfer of IBAD MgO technology to industry, and also demonstrates the utility of this technology for two very different YBCO deposition methods (PLD and MOD).
- * Renewed our CRADA with SuperPower, Inc. and signed a new CRADA with American Superconductor, both will support coated conductor activities in industry.
- * Engaged in sample and information exchanges with Oak Ridge, Argonne, and Brookhaven National Laboratories, leading to joint publications. Results from some of these collaborations are being reported this week in the Strategic Research Session.

Scoring criterion: Research Integration (continued)

- * Began a formal relationship with Lawrence Berkeley National Laboratory to draw on their long-standing expertise in IBAD with a PLD vapor source. The purpose is to analyze differences in films deposited with this technique in order to better understand the IBAD process in general. Initial results were reported by LBNL yesterday.
- * Fourteen of 25 Core Program publications since 2002 are the result of joint activities with national laboratories and companies, as well as six universities.

Scoring criterion: FY 2003 Results

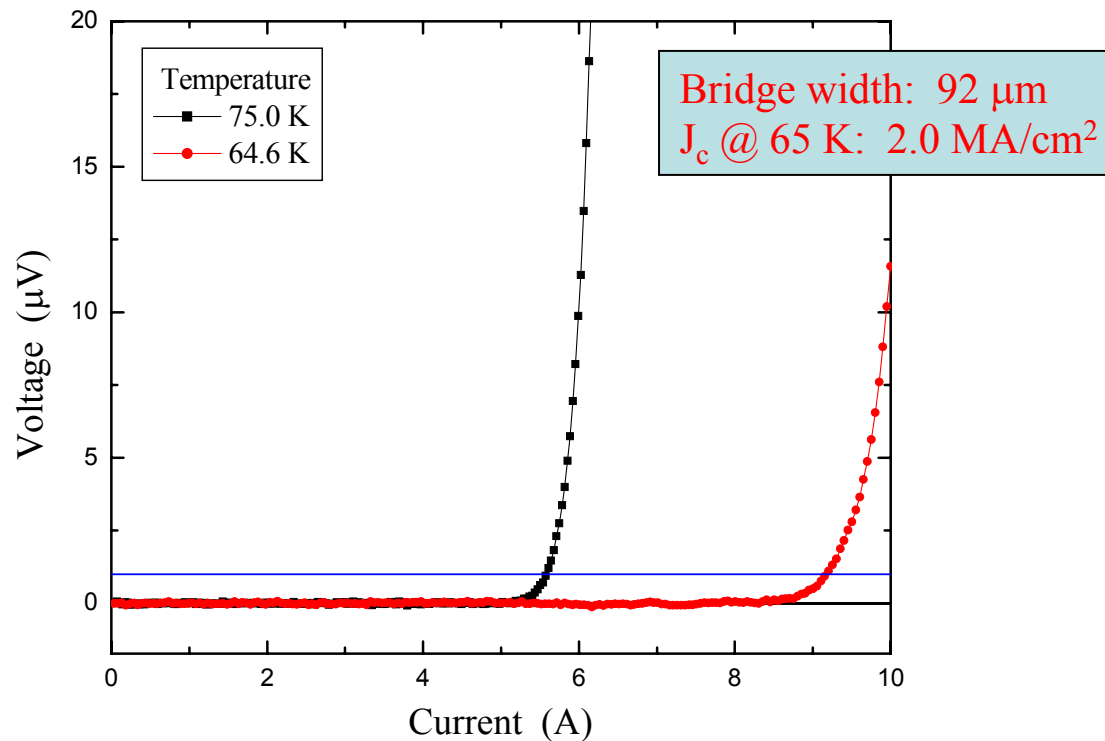
1. Improved the MgO texture on meter-long tapes to $\sim 7^\circ$ FWHM, enabling the deposition of YBCO with in-plane texture of less than 3° FWHM. *This resulted in J_c values exceeding those on our best IBAD YSZ and equal to those for single-crystal substrates.*
2. Analyzed diffusion of substrate elements into the YBCO and found that Al_2O_3 is an effective diffusion barrier. Also found that Er_2O_3 can be an effective barrier with the potential for replacing both Al_2O_3 and Y_2O_3 . *This will allow us to reduce the number of nonsuperconducting layers.*
3. Ion-milling experiments showed that thinned films and as-deposited films have the same J_c vs. thickness dependence. They also revealed no adverse effect of substrate elements diffusing into the YBCO, *suggesting that barrier and buffer layer thicknesses can be reduced.*

Scoring criterion: FY 2003 Results (continued)

4. Made a transition from SrRuO_3 to SrTiO_3 as the buffer layer between MgO and YBCO, with an improvement in performance. *STO is less expensive, more chemically stable, and can be fabricated into a more robust PLD target than SRO.*
5. Discovered that the porosity problems found in thick YBCO on IBAD YSZ do not exist with IBAD MgO; believe this is due to the smoother substrate surface used for the latter. *We no longer need the Y-Sm multilayers, described in previous years, for thick, high-current films.*
6. Produced many thick film bridge samples on IBAD MgO with $I_c > 300$ A/cm-width. *The best result is an I_c of 720 A/cm-width ($4.5 \mu\text{m}$, 1.6 MA/cm^2).*

Scoring criterion: FY 2003 Results (continued)

7. A 5 μm thick film – with a J_c of 1.2 MA/cm² @ 75 K – was measured in pumped liquid nitrogen. *Result: I_c (65 K) = 1000 A/cm-width.*



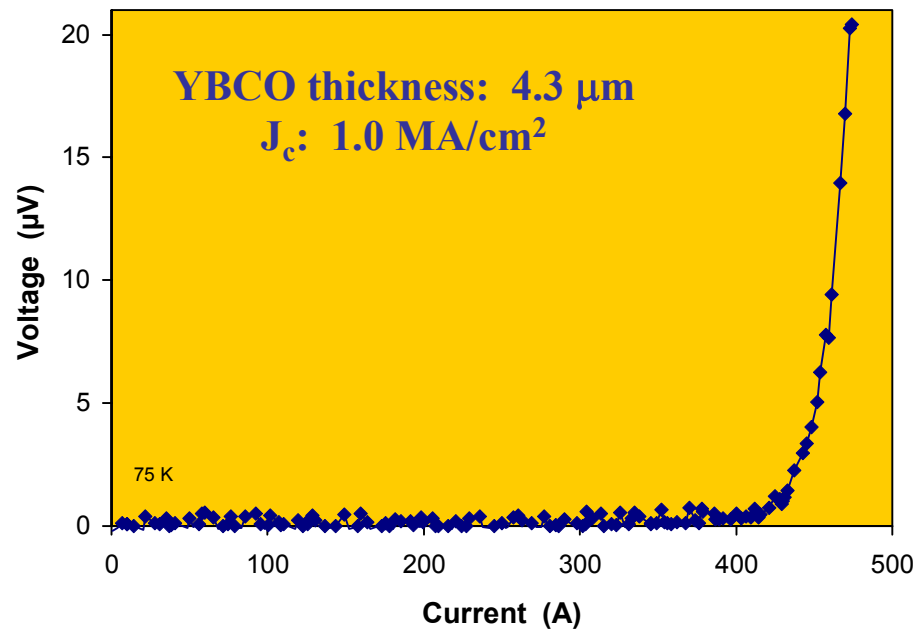
Scoring criterion: FY 2003 Results (continued)

8. Summary of cm-wide, continuously processed tape:
- ❖ Five 10-cm-long tapes with I_c s of 265-319 A, and J_c s of 0.9 – 2.2 MA/cm².
 - ❖ Two ~ 70-cm-long tapes with I_c s of 173 and 175 A.
 - ❖ A 54-cm section of one of the above tapes with $I_c = 233$ A.

Scoring criterion: FY 2003 Results (continued)

9. Deposited a thick film on IBAD MgO tape with the following result:

- ❖ 80 – 100 A at 0.6 T, $B||c$ over 14 cm
- ❖ Same tape, self field – 423 A over 1 cm



Scoring criterion: FY 2003 Results (continued)

10. Our industrial partners were successful in depositing high-current YBCO on Los Alamos-supplied IBAD MgO.
- ❖ American Superconductor reached 200 A/cm-width on small, batch processed samples with MOD YBCO. *This demonstrates that IBAD MgO is a viable coated conductor technology for ex-situ YBCO processing.*
 - ❖ SuperPower, Inc. reproducibly achieved 100-150 A on cm-wide, continuously processed tapes. *This is the latest accomplishment in the ongoing technology transfer of our IBAD and PLD processes to SuperPower through a CRADA.*

Scoring criterion: FY 2004 Plans

In FY 2004 the Core Program will substantially reduce its effort in the continuous processing of meter-long tapes, in order to focus on more basic and long-range issues that can impact both cost and performance of IBAD-based coated conductors.

- Experimentally investigate the fundamental texturing mechanisms of IBAD. *Goal: Develop a model that will allow us to further refine IBAD deposition parameters and texture.*
- Use extended I-V curves for IBAD MgO to evaluate the validity of the bicrystal plateau analogy for coated conductors. *Goal: Determine whether improved texture will yield higher J_c s.*

Scoring criterion: FY 2004 Plans (continued)

- Approach the drop in J_c with thickness as though it is a materials-processing issue, and not intrinsic. At a particular thickness, maximize J_c through a comprehensive process optimization. *Goal: Reproducible achievement of I_c s over 400 A/cm-width at a film thickness of $\leq 1.5 \mu\text{m}$.*
- Design and implement systematic experiments to determine if chemical modifications to REBCO offer enhanced performance, particularly in an external magnetic field. *Goal: Reproducibly double J_c at 75 K in a magnetic field parallel to the c-axis.*
- Use ion-milling and SIMS to determine the quantitative tolerance of YBCO to diffusing substrate materials, and use this information to minimize the nonsuperconducting layer thickness. *Goal: $< 100 \text{ nm}$.*

Scoring criterion: FY 2004 Plans (continued)

- Develop a single material that can serve as both a barrier and nucleation layer. *Goal: Reduce the number of layers by one, and reduce the barrier/nucleation layer thickness by 20 nm, with no reduction in performance.*
- Continue using our 1 meter tape capability to supply IBAD MgO to industrial partners and other national laboratories, and work with them individually to achieve optimum YBCO performance. *Goal: Three organizations depositing YBCO with J_c equivalent to those on single-crystal substrates.*

Conclusions

- IBAD MgO is an industrially-scalable process that involves a commercial alloy substrate, electropolishing, and less than 160 nm of deposited material between the metal and YBCO.
- From microbridges through meter lengths, we are achieving higher YBCO J_c s and higher cm-wide I_c s on IBAD MgO than we ever did on IBAD YSZ.
- We believe that there remain several areas in which the IBAD MgO structure can be simplified and superconductor performance on IBAD MgO can be enhanced – we will focus on these areas in FY 2004.
- The first step in transferring IBAD MgO technology to industry was very successful, and this transfer will continue next year.